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volume 3, Jan. 28 - Feb. 3, 1959.
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volume 5, July 29, 1959.

INDUSTRIAL RADIOACTIVE WASTE DISPOSAL

HEARINGS

BEFORE THE

SPECIAL SUBCOMMITTEE ON RADIATION

OF THE

JOINT COMMITTEE ON ATOMIC ENERGY

CONGRESS OF THE UNITED STATES

EIGHTY-SIXTH CONGRESS

FIRST SESSION

ON

INDUSTRIAL RADIOACTIVE WASTE DISPOSAL

JANUARY 28, 29, AND 30; FEBRUARY 2 AND 3, 1959

VOLUME 3

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NOTE

This is Volume 3 of four volumes of the record of "Hearings on Industrial Radioactive Waste Disposal." It covers the second half of the current waste disposal research and development program. It describes and discusses the handling, treatment, and proposed ultimate disposal systems for highly radioactive wastes that are primarily evolved from irradiated fuel reprocessing plants and discusses the future estimates and economics of disposal of radioactive wastes. It also includes a summary of environmental factors related to these estimates and a discussion of the industrial viewpoint of the radioactive waste problem. Volume 1 covers the portion of the hearings concerned with the origin and nature of various types of radioactive wastes evolved from nuclear energy activities and operations in use to manage these wastes at various AEC and other installations. Volume 2 deals with the first half of the current waste disposal research and development program. It describes and discusses the treatment and controlled dispersal of low and intermediate level wastes to the atmosphere, surface waterways, the ground, and the ocean and environmental investigations associated with this phase of the development program. Volume 4 discusses the activities of Federal and State governmental agencies in radioactive effluent control and administrative relationships between these agencies. The international aspects of the problem are also dealt with in this volume.

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INDUSTRIAL RADIOACTIVE WASTE DISPOSAL

FRIDAY, JANUARY 30, 1959

CONGRESS OF THE UNITED STATES,
SPECIAL SUBCOMMITTEE ON RADIATION,
JOINT COMMITTEE ON ATOMIC ENERGY,
Washington, D.C.

The special subcommittee of the Joint Committee on Atomic Energy met at 10 o'clock a.m., pursuant to recess, in room F–39, the Capitol. Hon. Chet Holifield, chairman of the special subcommittee, presiding.

Present: Representatives Holifield, Durham (chairman of the committee), Price, Van Zandt, Hosmer, and Bates; Senators Anderson, Dworshak, and Aiken.

Also present: James T. Ramey, executive director; David R. Toll, staff counsel; and Dr. Walton Rodger, technical adviser, Joint Committee on Atomic Energy.

Representative HOLIFIELD. The committee will be in order.

We apologize for the lack of space. We were unable to get the Old Supreme Court Chamber and this was the only room available, so we apologize to our witnesses and our visitors for the crowded conditions.

We will proceed until noon today and we will have to adjourn promptly at that time for an executive meeting of the full committee. Then we will come back into session again at 2 o'clock this afternoon and continue in the afternoon in order to take care of the additional witnesses.

Our first witness this morning is Mr. L. P. Hatch of Brookhaven, who will speak on high level wastes.

Will you please come forward, sir?

STATEMENT OF DR. L. P. HATCH,[1] OF BROOKHAVEN NATIONAL LABORATORY

Dr. HATCH. Mr. Chairman, we have several charts to be shown and the written version of the testimony, I might say I will not read it, but we will follow through it along with the charts.

[1] Date and place of birth: March 31, 1905; Islesboro, Maine.
Education: University of Maine, B.S. in civil engineering; Harvard University, M.S. in sanitary engineering.
Experience: Research fellow (fluid mechanics), Harvard University, 1930–35; assistant engineer on design of water purification and sewage disposal projects, S. M. Ellsworth Engineers, 1935–37; sanitary engineer, design of mechanical systems for control of toxic dusts and gases, Tennessee Valley Authority, 1937–41; design engineer on water purification and sewage disposal works, S. M. Ellsworth, Jackson and Moreland, 1941–43; captain-major, U.S. Army, Industrial Hygiene Laboratory, 1943–46; civilian technical director, Army Industrial Hygiene Laboratory, 1946–48; sanitary engineer (group leader), Nuclear Engineering Department, Brookhaven National Laboratory, 1948 to present.
Publications on the subjects of fluid flow through granular media, control of toxic dusts and gases, ultimate disposal of radioactive wastes.

Before we get into this, I would like to say just a bit about the background of our approach to this problem. We are, as you know, dealing at the moment with the high level wastes or those wastes that are in very high concentration of the fission products and in low volume. So we can talk about waste disposal processes which are more refined, perhaps, and in the nature of chemical processes themselves, more than we could with the low level and the high volume wastes.

Some 10 years ago at Brookhaven, we embarked on a program to seek a solution of this long-range disposal problem by means of fixation of the radioactive wastes in solid materials, in stable solids. There are several reasons why we chose this method of attack. One was that we could find many examples of this sort of thing in nature itself. For instance, if we had the waste material bound in stable solids of a mineral character, we have many examples to indicate that it would be retained for a very long time. In the natural clays we have examples of aluminum being a major constituent but the aluminum is not released from the clay over millions of years. So if we could put radioactive wastes in such a material, it could be considered to be permanently fixed in that solid form.

This doesn't mean necessarily that we would have disposed of the waste. Obviously, we have to do something with the solid, but there is a very good reason why it is in a much less hazardous form than if it was not in the soil. For instance, in the physiological relation to this sort of thing, materials which are not soluble, presumably, may actually be ingested and adsorbed in the human body and eliminated and do no permanent harm.

Strontium 90, if it were encased in a solid mineral particle, could pass through the human body, could be treated as a dust particle, and eliminated by the natural processes and not go to the bone as strontium commonly does.

There were other reasons why the fixation in solids did seem to us to be a very logical approach. Most anyone, I think, whether he was a scientists or not, would agree that solids represent the most stable form of matter and certainly the gases and liquids would be much less stable and, therefore, much more amenable to being dispersed in the natural environment.

Another reason is that the waste disposal problem is commonly discussed in terms of maximum allowable concentrations where we have responsible committees deciding that such a concentration is acceptable and more than this would be dangerous.

However, this problem, probably more than any other, requires an extra measure of precaution, and a safety factor way over and above the normal safety factors that are commonly used in handling hazardous material seems to be in order in this sort of problem. So with all of these things in mind, we adopted the method of fixation in solids as being the most logical and practical, and in the long run probably the cheapest, method of handling the disposal problem.

If we turn to the written part of the testimony, you will see at the beginning—the fixation of atomic wastes in solids is being developed at a number of laboratories and installations in this country and abroad. This was certainly not true 10 years ago and it has been an evolutionary process that this has come about. This is a very important factor because it means that the people in the laboratories

and installations that are dealing with the chemical reprocessing of atomic fuels, those that are responsible for the operations day by day, are giving serious thought to this method of disposal. So the chance of it becoming a successful and economic method is much greater.

At the end of the first paragraph—the first among these reasons why this is a logical approach is that the material, the atoms themselves, are chemically fixed within the framework of stable solids. This is an important point, but not one on which we should elaborate, except to say that if the material was encased in the solid and the atoms themselves were not bound as individual entities, it would not be the type of fixation that we mean. For instance, if you were to cut this material up, and if I might use an example, a piece of rock salt encased in lead, but if you cut the lead apart the rock salt would be soluble in water. However, if the atoms in the salt were actually fixed, attached to a stable solid in a chemical fashion, then if you cut it or ground it the atoms would not be released.

The existence of large quantities of high level radioactive wastes, such as would be produced in a major atomic power industry, would create a very special problem in that the amounts of long-lived materials at any one time would be sufficient to seriously contaminate very large regions of the earth for centuries to come. This statement is made simply to emphasize the fact that if we were to go on for 50 years in the atomic power industry, and find that we had reached an impasse, that we had been doing the wrong thing with the wastes and we would like to reconsider the disposal methods, it would be entirely too late, because the problem would exist and nothing could be done to change that fact, for the next, say 600 or a thousand years.

Representative HOLIFIELD. Would it be safe to say that this problem of permanent disposal of high level wastes must be solved and solved soon in order that these tremendous aggregations of contaminated material of high level power shall not become a public hazard throughout the centuries?

Dr. HATCH. Yes, I believe that is right and, in addition, this hearing and other such gatherings that serve to emphasize this point will be of first importance when we come to follow through in the large waste disposal operations that must follow if the atomic industry is to go ahead.

Representative HOLIFIELD. With the 15 or 17 reactors which are now being built, which will come on the line within the next 2 or 3 years, we are going to have a very rapid multiplication of the quantities of this material to be disposed, and we are told that we already have 52 million gallons that will continue in one place. It gives some idea of the magnitude of what may be in store for us as we spread out.

Dr. HATCH. This is true certainly, and we haven't yet reached the point where economic atomic power or the goal of economic atomic power is entirely achieved. When that does happen, however, there will be a much more rapid accumulation of wastes than may be foreseen today.

Representative HOLIFIELD. At the present time, this material is a liability. I know there is some thinking, but is there any practical development of a potential asset in this material, something which can

be taken from this waste and utilized extensively and which would reduce quantities, which is from a hazard standpoint.

Dr. HATCH. There is obviously that potential and there will be some testimony on that today. However, I think something should be said here along that line.

There is a great desire to think of waste products as being useful. This is a natural desire, of course. However, it sometimes is used as a substitute for the real disposal problem.

Representative HOLIFIELD. In other words, this is a hope not based on the historical utilization of wastes?

While waste has been utilized, it has never been utilized to the extent of removing the limit of waste from the economic picture?

Dr. HATCH. This is true. Dr. Wolman made this point the other day. The distinction between using the waste material for a purpose, which stands on its own merits, and thinking that this is also a means of solving the other problem of getting rid of it. These two things do not run hand in hand.

The other thing I would like to mention is that the thought of using atomic wastes for producing electric power in any large amount is unrealistic. The amount of energy involved in this material is very tiny.

Representative HOLIFIELD. In other words, the toxicity is great, but the energy content is small?

Dr. HATCH. The energy is great in the physiological sense. It does great harm because of its energetic particles, but to think of using this material to run electric lights and washing machines is just out of the question and has no meaning at all.

Representative HOLIFIELD. And this comes about because of the very small fraction of U–235 and other fissionable material in the natural uranium?

Dr. HATCH. The fact that the very nature of the energy itself is not conducive to thinking in terms of large amounts, doing large amounts of work with it.

Special applications for batteries and that sort of thing, such as we read about in the paper last week, this might be one of the rare cases where you deal with a very special form of energy, but to think of utilizing this material for power-producing purposes is certainly outside the scope of what we are thinking now.

Returning now to the written testimony, as we attempted to find a possible solution to this problem, 10 years ago, we also had to think in practical terms of how we would go about it. Again we looked to nature for an example. There are materials in nature that have very desirable properties for the problem in hand and again I cite the case of a natural clay, a special kind of clay. The reason that we selected this material is that it can enter into a reaction with the fission products, a chemical reaction, and cause the bonding of these two things together. It is very helpful to have this built-in chemical capacity in the clay material and after the waste products are incorporated in it we can heat the clay to a high temperature and seal them there so that they will not be disturbed and not be removed.

The first chart (fig. 1, p. 1842) shows a method of preparing the clay material in such a way that we can move the solution through it.

Representative HOLIFIELD. Could the Chair interrupt the witness for just a moment to note the presence of a new member on the Joint Committee on Atomic Energy, the distinguished gentleman from Vermont, Senator Aiken, who was appointed to this committee yesterday. I know it is going to be a matter of real pleasure to every member of this committee to have him as one of our members. We welcome you, sir.

Senator AIKEN. Thank you, Mr. Chairman.

Representative HOLIFIELD. We look forward to your other colleague who was also appointed yesterday, Senator Bennett.

Senator AIKEN. We will do the best we can to be helpful.

Representative HOLIFIELD. You always do the best you can and your best is much better than a lot of us.

Senator AIKEN. Thanks for saying so.

Representative HOLIFIELD. Thank you, Mr. Hatch, will you proceed please.

Dr. HATCH. As I say, this is a means of preparing the clay material such that we can use it in an industrial chemical process. The clay as we get it is a finely divided powder. There is no way that we could efficiently mix the liquid waste with the clay in the powder form and then separate them as we would have to do. So in this picture the clay has been mixed into a paste and extruded so that the waste solution can be passed through the column and the chemical reactions between the fission products and the clay take place in this manner. The chemical reaction that I speak of was mentioned yesterday briefly by Dr. Theis in speaking of waste moving through the soil, and the clay certainly is a common and a very important ingredient in soil. There is a so-called adsorption or an ion exchange process that goes on, and he also likened it, as I have in this paper, to the softening of water in which case certain atoms or ions in the liquid go into the solid phase. They attach themselves to a solid and other atoms or ions in the solid are given up to the liquid and we have an ion exchange, and the softening of water is a magnesium or calcium exchange for sodium. In this case it would be strontium and cesium particularly in exchange for ions in the clay, which would be calcium and magnesium. This sort of system lends itself well to the design and operation of a chemical plant and it makes for a convenient and simple way of handling the waste solutions. You can do it by remote control.

Representative HOLIFIELD. After the adsorption of the radioactive material into the clay occurs, do you further compress those clay cylinders into a solid mass?

Dr. HATCH. We haven't done that. This would be possible. We haven't done it for the reason that one likes to do as little as possible to materials after they become highly radioactive.

Representative HOLIFIELD. Is there then a natural flowoff of the liquid leaving practically a solid?

Dr. HATCH. Yes. We would expect that an amount of liquid equal to several hundred column volumes would move through the column before the clay capacity would be exhausted.

Representative HOLIFIELD. What is the efficiency of a given amount of liquid that goes through that, of the clay cylinder?

Dr. HATCH. That is a very important question and one that I am glad to try to answer.

The capacity of the clay—we will have to think in specific terms now—is very high. As for strontium, say, if we had a solution which contained nothing but strontium salt, we could put about 1 pound of strontium, in something like 20 pounds of clay, 1 in 20. I might say, in connection with this, I don't want to give the impression that we are going to achieve anything like this in practical operation, because the waste solutions are not pure. They contain many other things besides strontium, but the capacity of the clay is expressible in these terms. When we estimate that the clay is something like $50 a ton, we get some idea since we would be dealing with reasonable tonnage of this material, that the cost is not great. So the capacity of the clay is very large, shall we say, for what we want to accomplish.

The capacity that we are really interested in at the moment is more akin to how good a job we do in removing the waste material and this we will see in one of the later charts.

Representative HOLIFIELD. May I ask you one question? This clay adsorbs this on the basis of an exchange of ions and not on the basis of an adsorption like a sponge would absorb water, in addition thereto.

Dr. HATCH. That is also a very good point. That is exactly right and the distinction should be made strongly. This is in every sense of the word a chemical reaction.

Representative HOLIFIELD. It is not really an additional mass.

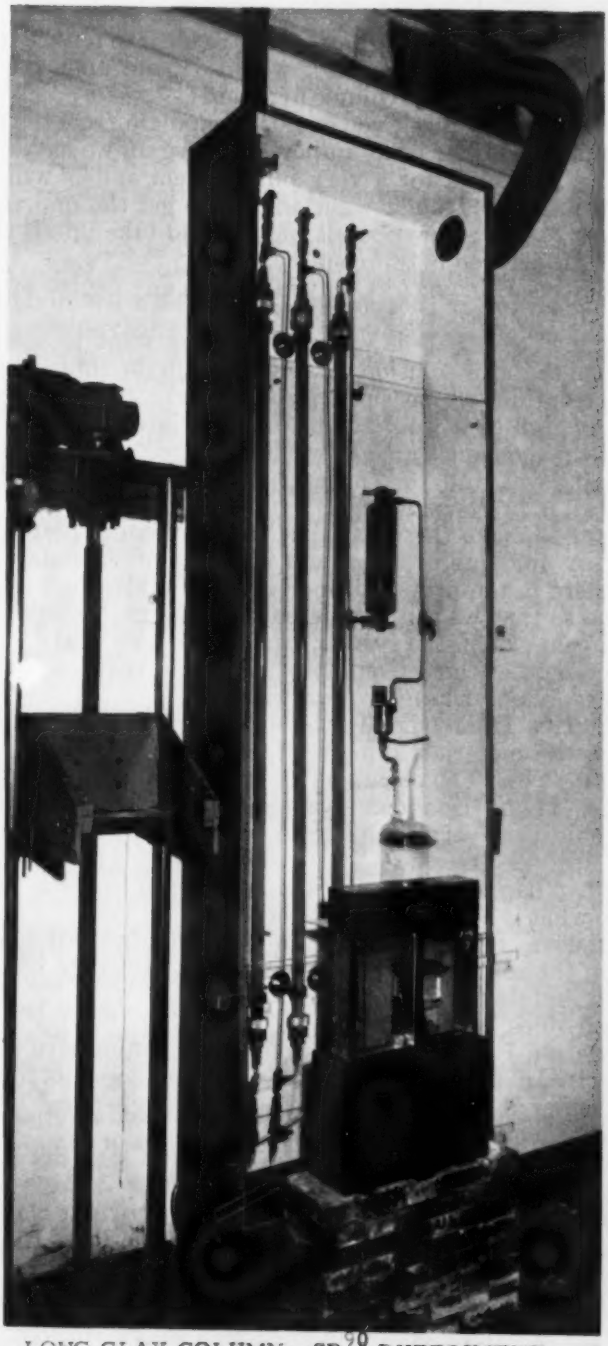
Dr. HATCH. That is right. It is not attached to the surface and usually these attachments are very weak. This is a chemical bond in itself and one which obeys and follows the standard laws of chemistry. So it is a very important distinction to make, and I think this goes a long way to helping in the final sealing of the material in the solid form.

This is a picture not shown in the written testimony. This shows three columns such as the one we just saw, the three here. They are glass tubes 5 feet long, about an inch in diameter, containing the clay, and they are connected in series and the solution can flow through the first, the second, and the third. This was done to measure what you just spoke of; the capacity of the clay to remove strontium, not so much with respect to how many pounds of strontium, but how much we purify or decontaminate the solution.

This is really the important thing because when the solution comes out, it has to be very highly refined. In high level waste treatment we must talk about purification in the order of 10 to the 8th; to really accomplish what we want to accomplish. If we reduce the concentration by a factor of a hundred or a thousand, the result is not worth very much to us.

The next chart, please.

Representative HOLIFIELD. Before you leave that, you have spoken of the adsorption by exchange of ions of 1 pound of strontium to 20 pounds of clay. I suppose it would be a variable factor as to the volume of liquid waste which would go through that type of an adsorption process. In other words, the 1 pound of strontium might be represented by many thousands of gallons of liquid waste that would have to flow through?



LONG CLAY COLUMN - SR^{90} EXPERIMENT

Dr. HATCH. Yes, the amount of water is immaterial.

Representative HOLIFIELD. So it would be hard to reduce that to understandable percentages because it would depend upon the intensity of the contamination of the volume of liquid?

Dr. HATCH. Yes. I think you could extrapolate from the statement that the volume of water is not important. Again we have to emphasize that the 1 to 20 is for a very pure system and it will never be achieved in reality. I don't want anyone to get the impression that we are going to use a few pounds of clay and take up all the fission products manufactured in this country.

Representative HOLIFIELD. Can you give us any degree of purity of the effluent that comes out after?

Dr. HATCH. Yes. This is exactly what we want to show. I am just reminded that this solution passing through the columns contained a level of strontium 90 measured by 2 curies in 10 gallons of liquid. That isn't as high as it would be in the high level wastes at Hanford perhaps, at the moment, but it is within a factor of something like 25 to 50 of that level, and for engineering study purposes this is high enough to call it full level wastes.

I just want to make the point that we are dealing here with more than just tiny amounts of strontium 90. The other materials in the waste are there in the proper proportions. They are not radioactive. This is done so that we can handle one radioactive isotope at a time, measure its behavior in the system, and not have to worry about what happens to the other things, but they have to be there so the chemical situation is representative of the real waste.

Representative HOLIFIELD. Is there concurrent adsorption of the other radioactive materials along with the clay?

Dr. HATCH. Yes. The chemistry of the system is, I am sure, a very complex one. If one tried to analyze exactly what happens to each one thing, it would be a very complex problem, but by using radioactive tracers, in this case strontium 90, we have a very great advantage in studying what happens to at least one of them without being concerned as to what is happening to the others.

Representative HOSMER. Do I understand that all your other chemicals in the solution tend to have an ion exchange and combine with the original clay? Is that correct?

Dr. HATCH. Yes.

Representative HOSMER. Whether they are radioactive or not?

Dr. HATCH. In all chemical processes, one does not distinguish between whether it is radioactive or not. The chemical behavior is not affected, in common terms at least.

Representative HOSMER. What I am trying to seek out is whether this is a general chemical combination, or does it just pertain to specific elements which will combine with this particular clay?

Dr. HATCH. No, it is specific in this sense—it pertains to positively charged ions. The negatively charged ions are not appreciably——

Representative HOSMER. No, sir; without a positive charged ion. Therefore, there is some analysis of your waste solutions necessary.

Dr. HATCH. There is. It is a very fortunate thing and this ties in also with the objective that the things that we are concerned about in the atomic wastes very heavily are these positively charged atoms,

strontium, cesium, and others. There is one notable exception to all this. It is ruthenium. You heard yesterday about ruthenium proceeding through the soil in some studies, I think at Oak Ridge, and the reason for this is probably the fact that it is not a positively charged ion, a cation.

Representative HOSMER. I am wondering if there are so many of the nonradioactive positive charged chemicals in waste that they pollute the system, and therefore you are taking up a lot of stuff that you don't need to take up.

Dr. HATCH. That is true. It is one of the real problems that we have to face and it goes back perhaps to the heart of all of this, the coordination of the waste disposal operations and the chemical operations that precede them. What the people do in the chemical processing plant has a great deal to do with what one has to do in the waste disposal plant. As I mentioned before, the trend toward systems which will fix this material in stable solids, which is now going through the minds of people in all the process plants, is a very important one in this regard. If they have the objective to fix the wastes in solids they will be able to do a great deal to arrange the situation beforehand.

Representative HOSMER. In other words, you may have to treat out some things from your solutions before you put them in there?

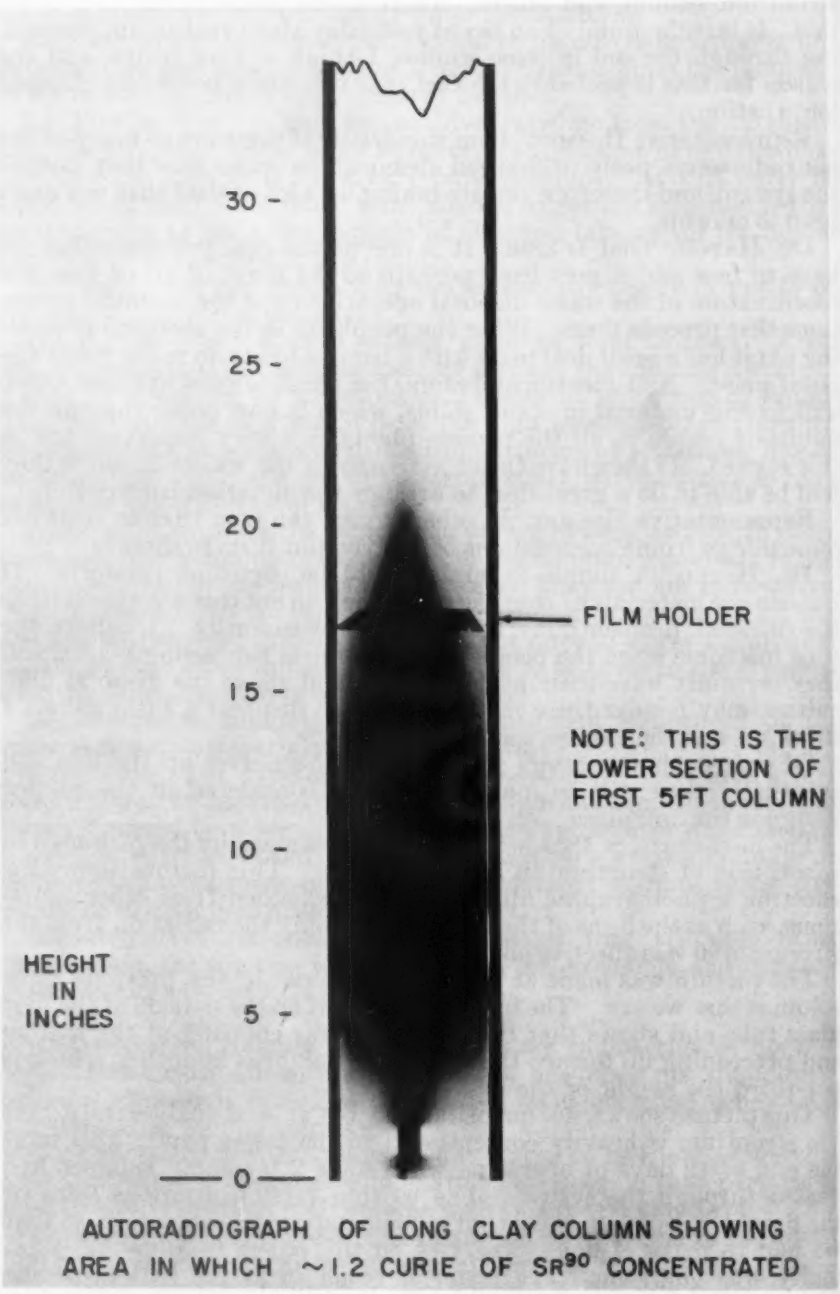
Dr. HATCH. A simple example would be corrosion products. If one can use materials of construction which do not corrode appreciably the disposal problem may be reduced tremendously. I believe the time has come when the people in the chemical processing plants, and they certainly have their problems over and above the disposal difficulties, may ponder these matters of waste disposal a little more. I think we will then achieve a great advantage.

Of course, these matters will all work themselves up the line and eventually they will be more thoroughly considered in the reactor design in the first place.

The next chart, p. 1834, will show what we mean by the pick-up, the adsorption of strontium in the clay system. This picture shows the effect on a photographic film strip. It is shielded from other radiations, such as the light of the room, so that only the radiation from the strontium 90 was effective on the film.

The picture was made at the end of the run of the previous three columns that we saw. The film strip was put on the outside of the first glass tube and shows that the strontium was entering at the bottom and proceeding up toward the top. This height is 20 inches, which is one-third the height of the first column.

This picture shows, not quantitatively but at least qualitatively, that the strontium is heavily concentrated in the lower part. This is at the end of 12 days of operation when some 200 column volumes had passed through the system. The written version mentions some of the figures. As I say, this is not a quantitative determination, so that we had to make chemical analyses on this waste to know what actually was going on. The material going in at the bottom of the column had the 2 curies in 10 gallons, which amounts to roughly 120 million strontium atoms disintegrating each minute in each milliliter of liquid, the milliliter being a cubic centimeter. Coming out at the top at the end of 12 days, the activity was being reduced to less than 1 disintegrating atom of strontium in this same volume. This calculates out to roughly 180 million reduction of strontium 90 in this system.



AUTORADIOGRAPH OF LONG CLAY COLUMN SHOWING AREA IN WHICH ~1.2 CURIE OF Sr^{90} CONCENTRATED

Representative HOLIFIELD. That is a very efficient elimination, isn't it?

Dr. HATCH. It is, except you then have to face a discouraging fact that you may need even more and probably do, so that reaching the level of a million reduction doesn't permit one to be elated in this sort of work, and we are really aiming at the hundred million or the billion reduction factor.

Representative HOSMER. Before you get away from that, you say this was circulated through there almost 200 times?

Dr. HATCH. It was. It didn't come back. It just kept on going.

Representative HOSMER. Oh, it was not recirculated?

Dr. HATCH. No, not at all. We don't want to recirculate it back.

Representative HOSMER. If it were recirculated, you shouldn't get that pattern on the film?

Dr. HATCH. You would tend to, but the recirculation would change the picture.

Representative HOSMER. It would diffuse it?

Dr. HATCH. Yes. I think one could safely say that up until the end of this time when this picture was taken, the situation was even better. Certainly it starts from zero and proceeds up to this point, so it is important to know that the material was passing through.

I think we could have the next chart now. Now we proceed to perhaps the most important aspect of the problem. When we think of the safety aspects of what we are trying to accomplish, we have to think of the end result. No matter how well we take up the strontium on the clay, it is of no great value to us for the 500- or 1,000-year period if we don't do more. The clay that we had in the previous column might be out anywhere in nature. The clay actually came from California. It is a special kind of clay and it is mined there. If one found this material in the soil as you will in lesser degrees in most places you could get a similar uptake of strontium on the soil. But in the long run I think this would not give the end result that we would want because, since the strontium would not be fixed there, another chemical solution could move it along, and these chemical solutions are not unheard of in the natural environment. For instance, hard water will readily flush this material on. Another thing, I won't try to go into any quantitative numbers, but it is important, is that the carbon dioxide in the air which absorbs in rainwater making carbonic acid can also move this material along. So everywhere in this country—Hanford and elsewhere—wastes trapped in the soil are subjected to things of this nature and one has to think of the 500-year period and what can happen in this way to flush this material on.

Representative HOLIFIELD. This means that you have put some kind of ceramic container or other type of hardening around this clay in order to keep it free from the oxidation of the air and other liquids?

Dr. HATCH. A little more than that. The fixation is within, and not without, and I think to define our objective more nearly than anything else, we want this containment to apply to the individual atoms themselves, so any container around the clay is not in our consideration for the moment. We fix it there by doing what you just said, making a ceramic. We do it by heating the whole mass. If the clay is heated to about 1,700° Fahrenheit the radioactive material will be sealed in the clay to a high degree. So that we had to devise a means of testing the clay to see how well it was sealed within, and this chart

(fig. 2, p. 1843) shows the result of a leaching test, a very simple test, by exposing the clay with the radioactive material in it to sea water. This is a high ionic strength solution and it is something nature has in great abundance and there would always be the possibility that it would come into contact with the solid. The chart shows the amounts of fission products removed by the liquid in terms of percentage of the amounts contained in the solid.

The chart does not cover the full length of our experiment but it goes to 4 years and we have data now up to 6 years. The numbers on the chart indicate that clay has been heated for a period of 8 hours, for 16 hours, for 48, 24, and 72 at the temperature. This is important, as you can see, because the 8-hour period shows that the radioactive material is progressively being released, at least up to 4 years. One cannot say whether or not this will level out in the future. It probably will not ever shoot up but will go on. As we come down the line, we have the 72-hour heating curve which shows not only by appearance but by actual chemical determination, that there was no progressive release beyond the first few weeks or days of the test.

Now in the percentage numbers, as you will note, this is one-tenth of 1 percent. The magnitude of the small number is not of any great consequence. This may represent a considerable amount of radioactive material, even the 0.08 percent. But usually the solid has some little contamination on the outside and you do not have a clean solid system. So we do for the moment simply convince ourselves that this amount of material is of no consequence, provided it is not released from the solid beyond that point.

Representative HOSMER. Those lines represent the accumulative amount of leach?

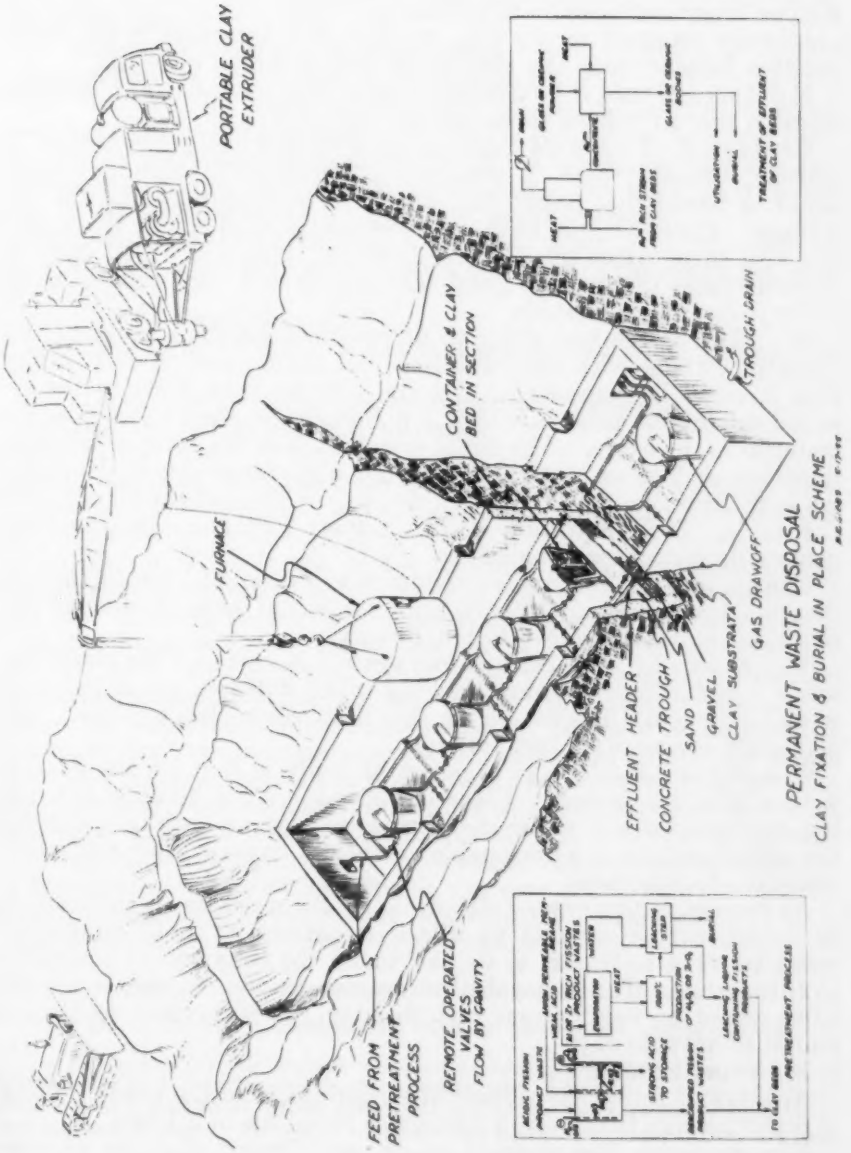
Dr. HATCH. Yes.

Representative HOSMER. In other words, on your 72-hour base, you have about 0.08 percent at the beginning and then there is none? It stops?

Dr. HATCH. Yes, that is the point. I might say that such a test, very simple though it is, involves a great deal of the meaning of what we are trying to accomplish. If the leaching were done, and we have done this on the very same material, using distilled water rather than sea water, we have this sort of straight line whether we had heated the clay for 8 hours, or 16, or 72 hours. Showing that the leaching test was not really a significant one.

I think perhaps we have gone through the clay fixation. The next chart (p. 1837) will show just one more thing. This is, as you see, an artist's conception of what a clay disposal plant might look like. Please don't feel that we have designed this plant as engineers should design things. This is just a representation of what we have in mind for the moment. This would represent a series of clay columns not contained in glass tubes obviously, but in durable containers about 5 feet in diameter and 5 or 6 feet tall. They would be constructed of refractory materials in order that one could heat the whole mass to the high temperature and not disturb the system at all, just leave it in place.

This represents a portable furnace on a crane to be lowered over the columns. The main design principle outside of the clay columns themselves is the arrangement of the columns such that we would get the best efficiencies out of the system, a so-called countercurrent operation. This drawing does show that the engineering feasibility of this is a matter which we have been considering all along.


PORTABLE CLAY EXTRUDER
CONTAINER & CLAY BED IN SECTION
TROUGH DRAIN
FURNACE
EFFLUENT HEADER
CONCRETE TROUGH
SAND
GRAVEL
CLAY SUBSTRATA
GAS DRAWOFF
REMOTE OPERATED VALVES FLOW BY GRAVITY
FEED FROM PRETREATMENT PROCESS
PERMANENT WASTE DISPOSAL
CLAY FIXATION & BURIAL IN PLACE SCHEME

Now we have to go back, if you will, sort of retrace our steps a bit to the matters that you gentlemen have been asking about, what other things in the waste do interfere with the absorption on the clay. There are many, particularly with respect to those wastes which are derived from the reprocessing of aluminum and zirconium fuels. These wastes obviously have large quantities of aluminum and zirconium ions, and these materials are positively charged ions and they are firmly attached to the clay, so that they interfere in the worst possible fashion with the absorption of the fission products.

Mr. TOLL. Would it be possible to pass it through another chemical solution first to remove those interfering ions?

Dr. HATCH. I am sure this is a question one could not answer in general, because there is always that possibility. Specific things could be found that might take out just the aluminum and the zirconium. However, you have to be very careful that you don't take out also some strontium. The refinement with which you have to separate these things is so great that you have a very great obstacle to try to do a thing like this.

We felt that the best thing to do was to treat the aluminum and zirconium along with the strontium and cesium and in such a way that it could be disposed of directly. So we have gone to the so-called calcination process, which is nothing more than converting the material to the oxide. The liquid systems are in the form of nitrates, fluorides, and if we put these through high temperature processes, we evaporate the water and then decompose the salts until we have the oxide. It is important in this respect that aluminum and zirconium do form very stable oxides.

The amount of bulk oxide product that we would have in the calcination process is much more than we would have if we had the strontium and cesium, absorbed in the clay.

This chart (fig. 3, p. 1845) shows again a leaching of the aluminum oxide. In this case we were leaching it not with sea water but with weak nitric acid. The reason for this is simply a chemical one. The acid gives us a more accelerated test.

One of the difficulties in this sort of study is that we are dealing with a 500-year period. There is no way we can have a 500-year test condensed to a few months of time, so that we have to accelerate the system so that in a reasonable number of days or years we have a measure of a long time.

As you see, in this case of strontium 90, the waste has been calcined, as I said, but before this we added strontium 90 to the simulated waste to serve as tracer, so we can watch the behavior of strontium in this system. The removal of strontium, as you can see, is not the 0.08 percent, as was the case with the clay, but 80 percent, or 85 in a matter of days, 60 days.

Is my time running out?

Anyway, it shows the release of strontium from the oxide. There is really no way that we can finish this thing up in a hurry.

Representative HOLIFIELD. I might say, Doctor, that all of your prepared statement will be in the record. However, I just asked the staff member who was the theorist who set up this witness list, with 15 minutes each.

Dr. HATCH. I am sorry. I am afraid I don't understand how one could cover such a subject in the time allotted. I will apologize for the time.

Representative HOLIFIELD. You don't need to apologize. I apologize for the setup of the witness list, and the short time available to the committee. It is a complete impossibility to cover this subject in the time that has been set up on the witness list, however high our hopes might have been.

Dr. HATCH. We can end up here quickly.

Representative HOLIFIELD. No, you go ahead.

Dr. HATCH. This shows the removal of the radioactive material from the solid, but the material removed is susceptible to going back into the clay system. It is now free of the aluminum and the zirconium ions. There is one more chart that shows the kind of plant that we would need in this kind of calcination operation.

Representative HOSMER. May I ask you, what is the purport of what you just said—that you can leach out some of the nonradioactive chemicals?

Dr. HATCH. Leach them out?

Representative HOSMER. Without leaching out your radioactive ones.

Dr. HATCH. No; nothing specific to radioactive material.

Representative HOSMER. All the same?

Dr. HATCH. Yes.

Representative HOSMER. I am trying to find out if leaching with a particular type of leach would take out a particular element and leave the others.

Dr. HATCH. That is one of the delicate questions. Some things might react specifically; yes. It would take a broad knowledge of this sort of thing to say, but in general we think that one soluble substance behaves pretty much like another. This simply shows a diagram of a calciner plant. This is the rotary kiln unit that Dr. Lieberman spoke of yesterday, very similar in principle to one that you might see in the manufacture of cement. It has to be designed, of course, with all the delicate controls to be sure that you do not create more of a hazard around the plant than you are correcting in what you are doing, because dust produced in this sort of operation would be a very serious hazard. The control of dust is probably the outstanding feature in the design of this system. I think that is sufficient.

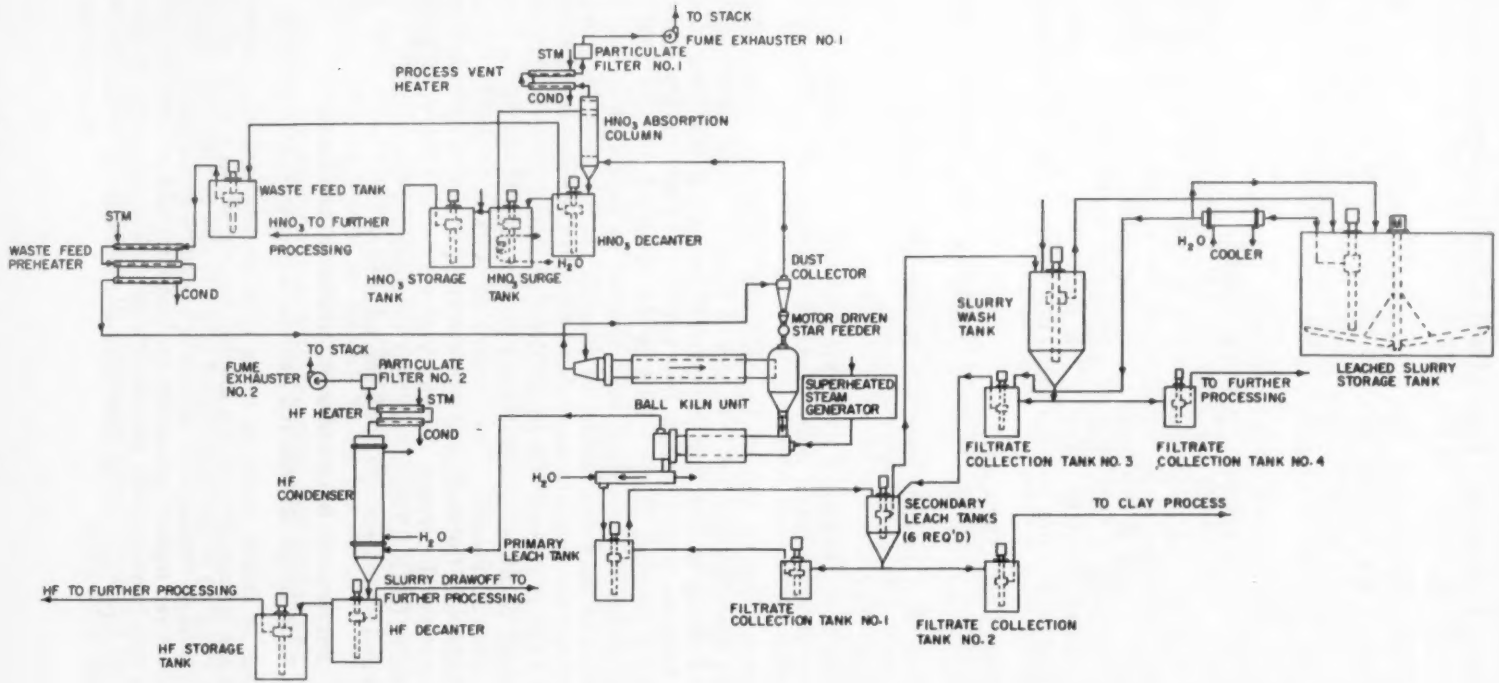
Representative HOLIFIELD. Thank you very much for your presentation. I think the information is very valuable for the record.

(The formal statement of Dr. Hatch follows:)

FIXATION OF RADIOACTIVE WASTES IN STABLE SOLIDS

(By Dr. L. P. Hatch, Brookhaven National Laboratory, January 1959)

The fixation of atomic wastes in solids is being developed at a number of laboratories and installations in this country and abroad as a basic and practical step in a method of ultimate disposal of the wastes by means of permanent and confined storage. There are a number of reasons why the fixation step represents a sound and logical approach to the disposal problem. First and foremost among these reasons is the fact that the method is designed to chemically immobilize the radioactive atoms themselves by sealing them within the framework of highly stable solids.



WASTE CALCINATION AND GAS RECOVERY SYSTEM SCHEMATIC

The existence of large quantities of high-level radioactive wastes, such as would be produced in a major atomic power industry, would create a very special problem in that the amounts of long lived atoms (isotopes), at any one time, would be sufficient to seriously contaminate very large regions of the earth for centuries to come. It is an elementary but highly important aspect of the long-range disposal problem that no physical, chemical, or biological changes in any way reduce the toxicity of radioactive materials and they become less hazardous only as they are reduced in quantity through the independent and time-consuming process of radioactive decay. For some of the fission products, strontium 90 and cesium 137, the decay process is extremely slow and these materials, once released in quantity to the natural environment, would pose a problem of the deepest significance. The reported appearance of the long-lived fission product, strontium 90, in food and water supplies, in vegetation and in soil, over vast areas of the earth, as the result of the testing of atomic weapons, should serve as a very forceful reminder in this regard.

It is with the foregoing points in mind that various methods of fixing the waste fission products in stable solids are being investigated as a basic preventive measure to insure that they would not at some future time be released to the natural environment either by accident, or by miscalculation, or by design.

At Brookhaven National Laboratory we have been working for some years on a process for ultimate waste disposal which involves the adsorption and fixation of the fission products in one of the natural clay minerals. Briefly, the system under investigation is based on the use of columns of extruded clay such as the one shown in figure 1. As the waste solution is passed through the column a chemical reaction takes place which results in an exchange of atoms (positive ions) between the liquid and the solid. The purpose is to bring about the adsorption of the radioactive atoms in the solid in return for a release of equivalent amounts of nonradioactive atoms to the liquid. The adsorption process, which is known as ion exchange, is analogous to the mechanism of softening hard waters by the exchange of calcium and magnesium ions in the hard water in return for sodium ions in a solid such as one of the natural zeolites, or the more modern synthetic ion exchange resins. As will be explained later, there are a number of other chemical substances in the wastes which, as ionic constituents, also tend to take part in the exchange process. As a result there is a basic competition between the fission product ions and the so-called bulk ions with respect to adsorption on the clay mineral.

The next chart (see p. 1831) shows an experimental system containing a series of three columns of clay which was used to measure the uptake, or adsorption, of strontium from a simulated waste solution. In this experiment roughly 10 gallons of the simulated waste which contained also amounts of strontium 90, as a tracer, equivalent to 2 curies of radioactivity (roughly 120 million atoms disintegrating per minute in each milliliter) was passed through the columns for a period of 12 days. At the end of the experiment the solution emerging from the system was found to contain strontium 90 in a concentration measured by less than 1 atom disintegrating per minute per milliliter. The net result was that the concentration of strontium (both radioactive and nonradioactive) in the waste solution at the end of 12 days of continuous operation was being reduced by a factor of about 180 million. The uptake of strontium 90 on the clay at various positions up the first of the three columns (in the direction of liquid flow) is indicated by the degree of blackening of a photographic film strip held against the outside of the glass tube containing the clay, as shown in the next chart.

The actual fixation of the fission products in the clay mineral, once it is adsorbed there, is accomplished by heating the clay to about 1,700° F. The term "fixation" as used in the present sense is but a relative term and becomes meaningful only when expressed in terms of quantitative values. We have adopted a method for measuring the degree of fixation by determining the amounts of fission products released by the fired clay, in which they are contained, to a leach solution which, as selected for our purposes, is ordinary sea water. The reason for this selection was that sea water would represent one of the strongest ionic solutions that might commonly be encountered in the natural environment and, therefore, the leaching test would be a rather severe one. The results of one series of measurements to determine fixation as a result of fusing mixed fission products in clay are shown graphically in the next chart (fig. 2). Here we concluded that there is no evidence of release of any significant amounts of the fission products to the sea water during a period of

FIGURE 1.

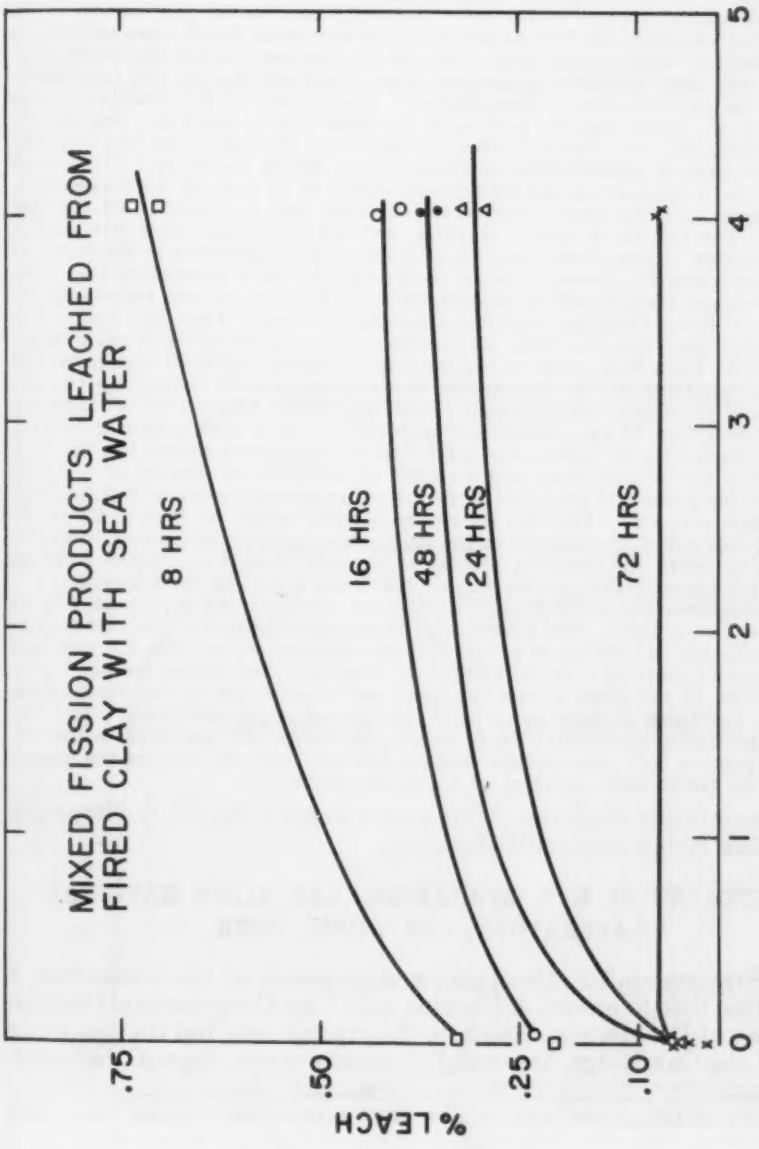

MIXED FISSION PRODUCTS LEACHED FROM
FIRED CLAY WITH SEA WATER
8 HRS
16 HRS
48 HRS
24 HRS
72 HRS
% LEACH
.75
.50
.25
.10
0
1
2
3
4
5
YEARS

FIGURE 2.

contact of more than 5½ years. At the very beginning of the experiment an amount of material representing some 0.08 percent of the total radioactivity was released to the liquid but we are inclined to attribute this to original surface contamination.

The actual waste solutions, as they emerge from the reprocessing of nuclear reactor fuels, contain other chemical substances which have nothing to do with the fission process itself but which, as ionic constituents, interfere with the adsorption of the fission products on the clay. Examples of such materials are aluminum and zirconium, which were originally used in the fabrication of the reactor fuel elements, as well as certain acids and salts which were introduced in the chemical reprocessing steps. In our development studies the wastes containing bulk salts of metals, aluminum, zirconium, and the like are calcined to produce the stable oxides of these metals. The purpose in the calcination step is, first of all, to convert the bulk salts to stable oxides which are suitable for ultimate disposal, after some further treatment, on the basis that they will undoubtedly contain considerable amounts of the fission products. The second objective is to bring about the separation, wholly or in part, of the fission products that do not form stable oxides, so that they may be adsorbed on the clay mineral. The results of typical leaching tests on aluminum oxide produced in the calcination of simulated aluminum nitrate wastes are shown in figure 3. In each test the simulated waste solution contained very small amounts of the radioactive isotopes cesium 137 or strontium 90 in addition to the representative amounts of these elements in the nonradioactive form. Thus the radioactive isotopes are used, one at a time, as tracers to show the behavior of the element in question. Data from tests on the leaching of mixed oxides of aluminum and zirconium produced in the calcination of simulated wastes containing the zirconium and aluminum fluorides and aluminum nitrate also spiked with cesium 137 and strontium 90 are represented in figure 4. As is seen in figures 3 and 4, complete separation of cesium and strontium from the bulk oxides has not been achieved in any of our tests, and we must be prepared to dispose of the bulk oxides on the basis that they will be permanently contaminated with these long-lived fission products. For this reason, as already noted, we look to the qualities of stability and permanence in the bulk oxides themselves, and we recognize the need to carry the leaching process to its conclusion from which point no further release of fission products from the solids would be forthcoming.

A drawing showing the outlines of a proposed calciner plant is presented in the next chart (see p. 1840). The basic design feature in this plant is that rotary kilns are used for the calcination step and that the gases emerging from the calciners would be condensed to a liquid insofar as possible. The reason we emphasize condensation of the gases is that the small amounts of radioactive dusts which would be entrained in these gases would be trapped in the condenser. The control of the highly radioactive dusts which, inevitably, are produced in the calcination process is a very serious problem and one that will require the utmost care in the design and operation of a calciner plant.

Representative HOLIFIELD. Our next witness is Mr. E. G. Struxness of the Oak Ridge National Laboratory.

STATEMENT OF E. G. STRUXNESS,[2] OAK RIDGE NATIONAL LABORATORY, OAK RIDGE, TENN.

Mr. STRUXNESS. Mr. Chairman and members of the committee, I would like first to answer a question asked by Congressman Durham yesterday of Dr. Morgan regarding the capital costs and the operating costs of the Oak Ridge National Laboratory waste disposal facilities.

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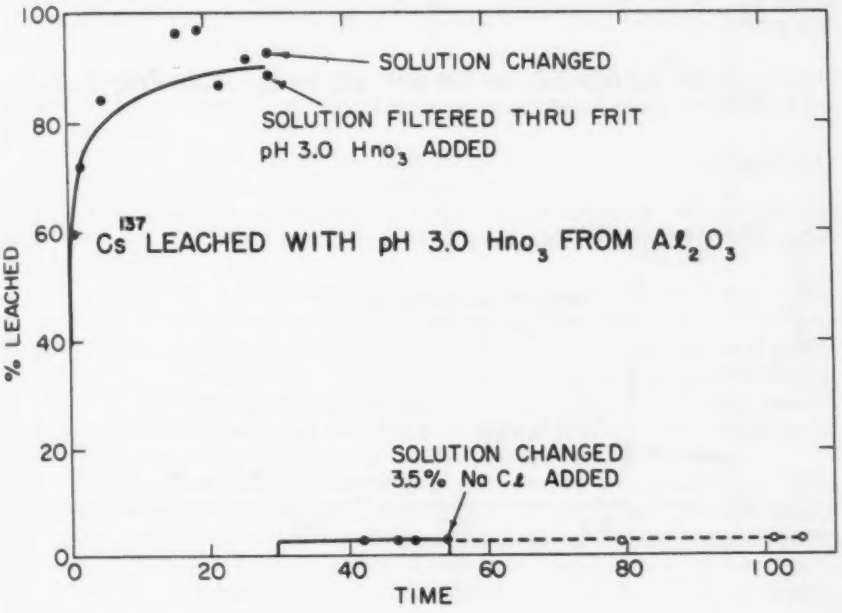
[2] Date of birth December 26, 1913, Nome, N. Dak.; Luther College, Decorah, Iowa, 1931–35, B.A., zoology, chemistry; Northwestern University 1940–42, field zoology, animal physiology; University of Tennessee, 1948–51, radiobiology; Oak Ridge National Laboratory, 1953–present, section leader; Union Carbide Nuclear Co., 1947–53, department superintendent; Carbide & Carbon Chemical Co., 1946–47, technical staff; Tennessee Eastman Corp., 1944–46, special hazards engineer; AAF technical training command, 1942–44, senior instructor; Senior High School, Blooming Prairie, Minn., 1935–40, science instructor.


100
80
60
40
20
0
% LEACHED
SOLUTION CHANGED
SOLUTION FILTERED THRU FRIT
pH 3.0 Hno3 ADDED
Cs137 LEACHED WITH pH 3.0 Hno3 FROM Aℓ2O3
SOLUTION CHANGED
3.5% Na Cℓ ADDED
0
20
40
60
80
100
TIME

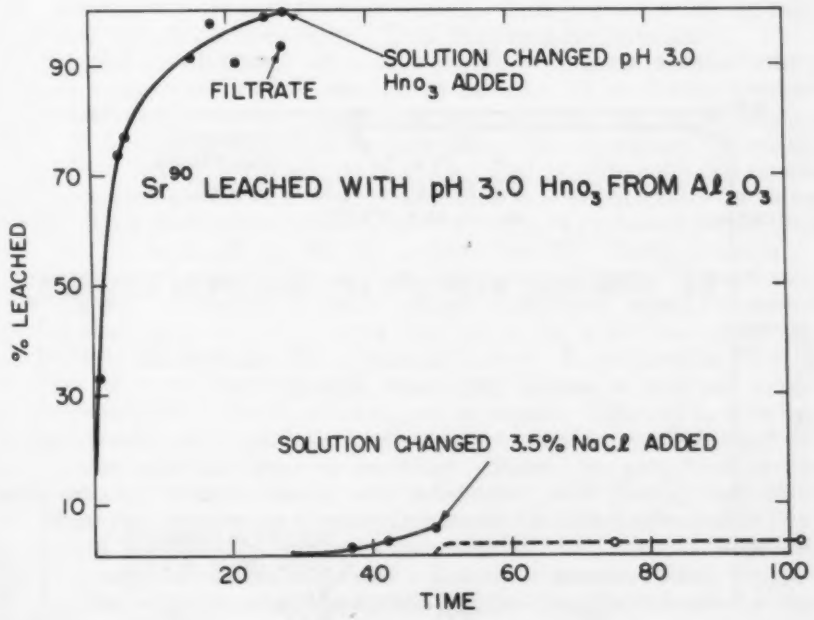

90
70
50
30
10
% LEACHED
FILTRATE
SOLUTION CHANGED pH 3.0
Hno3 ADDED
Sr90 LEACHED WITH pH 3.0 Hno3 FROM Aℓ2O3
SOLUTION CHANGED 3.5% NaCℓ ADDED
20
40
60
80
100
TIME

FIGURE 3.

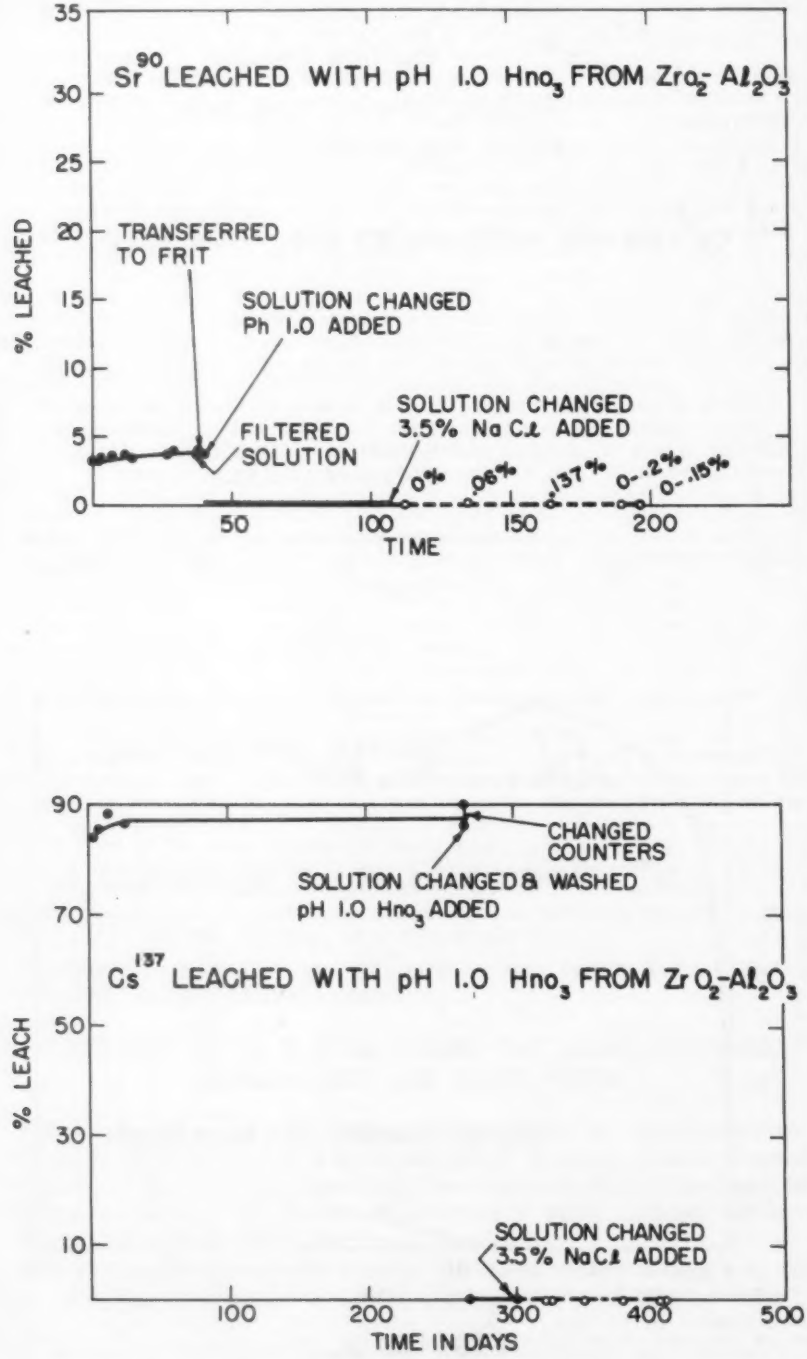

Sr 90 LEACHED WITH pH 1.0 Hno3 FROM Zro2- Al2O3
% LEACHED
TRANSFERRED TO FRIT
SOLUTION CHANGED Ph 1.0 ADDED
FILTERED SOLUTION
SOLUTION CHANGED 3.5% Na Cl ADDED
0%
.06%
.137%
0-.2%
0-.15%
TIME
Cs 137 LEACHED WITH pH 1.0 Hno3 FROM Zr O2-Al2O3
% LEACH
CHANGED COUNTERS
SOLUTION CHANGED & WASHED pH 1.0 Hno3 ADDED
SOLUTION CHANGED 3.5% NaCl ADDED
TIME IN DAYS

FIGURE 4.

The capital investment of the complete waste handling and disposal system is $2.7 million. The operating costs of these facilities runs approximately $269,000 a year.

The construction cost of the process waste water treatment plant, which is the question that Mr. Durham was specifically interested in, was $260,000.

Preliminary estimates of the cost of this lime-soda ash treatment is of the order of 18 cents per thousand gallons.

This testimony is contained in a document entitled "Fixation and Ultimate Disposal", copies of which have been submitted to the committee and distributed.

The possibility of converting radioactive liquid waste into an insoluble solid is appealing because it would free the high-level waste dispsal problem from its present dependence on the geology and geography of the disposal site. Even concentrating the radioactive fission products from liquid waste into solid form, leachable or not, would be a major step forward. Once they are robbed of the mobility of liquid flow, the fission products are far less hazardous.

In theory, many possible methods of fixation are available. The investigations at Oak Ridge have been limited to mixing the waste with clay and certain common fluxes and heating the mixture to form a brick or sinter. In particular, these investigations have been concerned with the possibility of using heat from the decay of the fission products in the waste to form a sinter.

Representative HOSMER. Would you further define the term "sinter"?

Mr. STRUXNESS. It is an overfired brick. This process would be self-contained and, therefore, safer. Two problems are involved:

First, the development of a mix which will form a satisfactory solid at a temperature low enough to be reached by fission product decay-heat; and, second, the design and engineering developments which will convert a laboratory procedure into a routine operation.

The first sintering experiments at Oak Ridge were centered on the use of Conasauga shale, a local geologic formation, literally as cheap as dirt. This shale contains a fair amount of a clay mineral called illite that has this high affinity for the cations that Dr. Hatch spoke of.

Preliminary tests with tracer quantities of radioactivity were very encouraging. Later tests with actual high-level waste, however, showed that some of the sintered cakes were far more leachable than we had been led to expect from our early work. In particular, Mix 15, the best of some two hundred waste-clay mixes, would not retain cesium when the sintered mix was put in water. This led to a period of considerable reevaluation, particularly as to the practical need for making the solid desistant to leaching. Should we plan to store the solid where it would remain dry indefinitely and put all our effort into obtaining maximum physical strength and mechanical stability? Or should we design a sinter so unleachable that it could be stored safely in any convenient location? For the present at least, we have decided to continue working toward our original goal of a leach-resistant solid.

The leachability of a sinter depends on several properties, including surface area and solubility. A porous or powdery sinter exposes more surface area to be attacked by water than one which is dense

and impermeable. The differences in the physical properties of a sinter depend on composition, particle size, degree of mixing, rate of heating, and so on. The art of baking "mud pies" so that they will always be dense and impermeable is one that we are still learning.

A more fundamental factor in leachability is the way in which the various radioactive elements are held in the solid material. The sinters are composed largely of very fine grained crystals, invisible by optical means but easily identified by X-ray diffraction analysis. Like all crystals, they consist of a lattice, or network; the snowflake is a common example of a type of crystal structure. We find that some of the radioactive elements are held mechanically in the openings of the crystal lattice, from which position they are easily removed. Other elements, however, are actually built into the crystal lattice itself and become part of it; these elements are not leached out easily. It is probably impossible to find a mixture which will build all of the fission products into the crystal lattice. For this reason, mechanical retention within the sinter must also be relied upon. Operational limitations make it desirable to form the fired sinter at as low a temperature as possible.

We are having some success with our search for a mix which will yield better crystals. For example, the addition of boron in the form of boric acid has greatly improved the retention of cesium, and we are studying changes which will improve the retention of strontium.

The engineering design of the pilot plant in which we hope to make a practical test with substantial amounts of radioactivity is based on our desire to use the fission product decay-heat to fire the sinter. This is not to save the cost of the power required, but because we feel the job can be done better this way. In the first place, the equipment is simpler and the combination of unbroken insulation and shielding around the sides and bottom of the sintering furnace provides a desirable element of safety. Heaters immersed in the sinter must have connections with the outside and these connections, in addition to the heaters themselves, are likely to fail. The heat from radioactive decay is not subject to failure. The cake, after it starts to harden, is a poor conductor of heat, and heaters imbedded in the cake tend to overheat the material nearest to them. Overheating can be serious because the off-gas problem is greatly increased by the volatilization of some fission products if even a small part of the mix is heated above the minimum temperature required for sintering.

The design of the pilot plant for self-sintering encountered a heat problem which we feel we have solved. The heat losses per unit volume are greater with a small furnace than with a large one because the surface area of the small furnace is large in proportion to its volume. There is a minimum size below which sintering temperature, $\equiv 850°$ C. (1562° F.), cannot be reached because of the practical limitations of thermal insulation.

The upper limit on size is determined by the hazards involved in handling large volumes of radioactive waste at our particular plant site. The compromise has been to build a small furnace surrounded by a heat barrier. The heat barrier is a blanket of exterior heating coils which will keep the furnace wall almost as hot as the sinter inside of the furnace, and so prevent heat loss.

Figure 1 (p. 1854) is a schematic drawing of this plant. It shows the biological shield, the mixer, the furnace, and the two containment vessels. Both the mixer and the furnace require a very efficient air cleaning system not shown on the diagram. During mixing there is some evolution of CO_2 which is conducted down this line and into the air-cleaning system. The air-cleaning system consists of a series of devices, a caustic scrubbing tower, followed by a condensate storage tank, a circulating caustic absorber, and a mechanical filter consisting of successive beds of sand, soda lime, and charcoal.

This air-cleaning system is so arranged that we can evaluate several combinations and cam sample at many points. The characterization of off gases and the evaluation of this system are important objectives in this experiment. In operation the liquid is jettisoned into the mixer through this line. The solid materials are put into the hopper and pass down this line into the mixer.

Following mixing, this valve is opened and the slurry is dumped down into the furnace.

Representative HOSMER. This system requires a very high concentration of your liquids?

Mr. STRUXNESS. Of the fission products; yes, indeed.

Representative HOSMER. In other words, you have treated your waste down to where you have practically what you want to go into that batch at the time it goes in. In the previous process described, as I understand it, you take less concentrate and keep running it through.

Mr. STRUXNESS. In the actual case, Mr. Hosmer, we do not plan to pretreat the waste. We simply plan to take the raw, "hot" wastes and introduce them into the system.

Representative HOLIFIELD. How do you get rid of the liquid? Through the evaporation?

Mr. STRUXNESS. A series of events occur in this sintering process:

First, the temperature is brought to boiling. Then the water vaporizes and is passed into the air-cleaning system. Then the acid aluminum nitrate itself decomposes and creates oxides of nitrogen gas which are passed into the air-cleaning system; then the temperature of the dry cake gradually rises to the predetermined sintering temperature.

Representative HOLIFIELD. How do you remove the cake after it has had its firing?

Mr. STRUXNESS. After the sintering temperature has been attained, we plan to turn off the barrier heaters and let the cake cool down; if need be, to remove the thermal installation around the furnace to speed up the cooling-down process.

Then, when the temperature of the cake has dropped to ambient temperature, which will take over a year and maybe several years, it is our intention to introduce water into the furnace and periodically analyze the water.

Representative HOLIFIELD. Does this not tie up your furnace for a long time?

Mr. STRUXNESS. No; it is designed in such a way that we can remove the mixer and, with the aid of lifting hooks on the cover, remove the complete furnace unit. The furnace is actually a double-lined furnace. The inner liner is attached to the cover.

Representative HOLIFIELD. Once you remove that cake from it, that has to be shielded until it cools down?

Mr. STRUXNESS. Indeed it does. It has to be shielded after it is cooled down, sir.

Representative HOLIFIELD. It takes about a year for the thermal heat to cool?

Mr. STRUXNESS. Yes.

Representative HOLIFIELD. But at the end of that time it is still very heavily radioactive?

Mr. STRUXNESS. Yes.

Representative HOLIFIELD. So in storage you would have some kind of shield, would you not, certainly in the transportation of it in place of permanent storage?

Mr. STRUXNESS. We have given some thought, following the removal of the mixer, to installing a core-drilling rig on the same supports and taking core samples of the cake in the furnace; that is, just withdrawing cores of the cake for further study.

Representative HOSMER. This is all experiment now. You are not talking about process?

Mr. STRUXNESS. That is right. This is just an experiment.

Mr. TOLL. Would it be an oversimplification to say that your process is primarily a baking or a drying-out process, whereas Dr. Hatch was talking about a chemical combination process?

Mr. STRUXNESS. Well, there is chemical combination process going on in this mix as well as in Dr. Hatch's procedure. We are concerned primarily about tying up cesium and strontium in the illite in the mixture.

Representative HOLIFIELD. After you imbed the radioactive material in the clay under the previous process you still have to have this firing in the sinter; do you not?

Mr. STRUXNESS. I am sorry, I didn't understand the question.

Representative HOLIFIELD. After the so-called flow process through the clay particles and the ion exchange which imbeds the radioactive material in the clay itself, then you have to take the clay, do you not, and bake it in order to sinter it?

Mr. STRUXNESS. Yes.

Representative HOLIFIELD. So this is a continuation of his process; is it not? It is not an ultimate process; is it?

Mr. STRUXNESS. Following the mixing of these ingredients, the heat from the fission products contained in the slurry will bring the temperature of the slurry to boiling, the water is driven off, the unstable salts are decomposed and finally a "fixing" temperature is reached.

Representative HOLIFIELD. So this is a continuation of the other process, of the flow of the mass of liquid through the clay particles?

Mr. STRUXNESS. It is similar in this respect, sir; that we rely on the ion-exchange capacity of one of the raw materials in the mix, namely, illite, to tie up the cesium and the strontium.

Representative HOLIFIELD. I understand that. Maybe my question isn't a good question, but after it is tied up, then you run it through the top and you tie it up. Then is it deposited in this cooking vat?

Mr. STRUXNESS. Yes. It simply sinters in the furnace. The one

remaining step in the complete process is the final disposition of the sinter.

Representative HOLIFIELD. The removal and the permanent storage?

Mr. STRUXNESS. Right.

Representative HOSMER. I am sorry to ask again, but Dr. Hatch mentioned that he had just slight traces of strontium in his test solution. He pumped this through these clays and the ionization process occurred and then he took the stuff out and baked it. Apparently he put through a high volume of contaminated fluid that he was trying to purify.

What you do in this is, you have a low volume of high concentrated fluid? In other words, before you get into this, you have concentrated your limits that you want to look into this sinter, and much the same as mixing cement, you mix them and then you have the sintering process generated from the radioactive heat itself rather than in separate furnaces?

Mr. STRUXNESS. Right.

Representative HOSMER. So you have to treat the waste chemically before it goes into this thing to a point where you have a very high concentration of whatever stuff you want to locate?

Mr. STRUXNESS. Actually, Mr. Hosmer, the scale of this experiment is designed in such a way that we can dissolve one MTR fuel or one ORR fuel element and pour that liquid into our mixer. The only treatment involved in this procedure is simply to mix the dry ingredients with the liquid waste.

Representative HOSMER. You are going to end up that way then with a much greater volume of the sinters than Dr. Hatch, is that right?

Mr. STRUXNESS. Well, we assumed that we could dissolve this fuel element and end up with about 30 gallons of very high level waste containing approximately 100,000 curies. We would let that cool for some period of time and then introduce it into the mixer.

Following the sintering procedure when the water is driven off and the unstable salts are decomposed and so on, we expect to end up with the radioactivity tied up in a cake about 8 inches thick and 24 inches in diameter. The volume of the final cake is about, as I recall, seven-tenths of the original volume of the liquid waste.

Representative HOSMER. You have, say, stainless steel. You have all that steel and this has to go some place. Where does this go?

Mr. STRUXNESS. In the actual case, were this procedure ever adopted as a routine disposal operation, these fuel elements would be dissolved. The unspent fissionable material would be separated from the fission products.

Representative HOSMER. That is actually done?

Mr. STRUXNESS. Yes. In the actual case, sir, we would take the wastes that come from the fuel reprocessing plant and introduce them into our mixer. The pilot plant is now 90 percent complete and we are awaiting the completion of our laboratory work on sinter development to make the initial run.

We cannot at this time foresee all of the engineering developments which might lead to self-sintering as a feasible and economical method of ultimate disposal. Undoubtedly we will continue to store these high-level liquid wastes for some time to come. For the present, tank

storage is the safest and possibly the cheapest method of containing the wastes. However, we do not regard containment of radioactive liquids in tanks as an answer to the problem of permanent disposal.

For reasons of safety, it would be better to self-concentrate the liquid to a solid residue in the bottom of the tank. Even this residue would be subject to decomposition and possibly too soluble to store there indefinitely.

When these factors are considered, it would appear even better to develop a method by which wastes in the storage tank would self-sinter to produce an insoluble ceramic in which the long-lived fission products are permanently fixed. Fixation and storage in tanks provides safe containment during the preliminary cooling period and has the added advantage of stability and, hence, greater safety for indefinite storage *in situ*.

Representative HOLIFIELD. Are there any further questions?

Thank you very much, Dr. Struxness.

(The formal statement of Mr. Struxness follows:)

FIXATION AND ULTIMATE DISPOSAL

(By E. G. Struxness, Oak Ridge National Laboratory, operated by Union Carbide Corp., for U.S. Atomic Energy Commission, Oak Ridge, Tenn.)

The possibility of converting radioactive liquid waste into an insoluble solid is appealing because it would free the high-level waste disposal problem from its present dependence on the geology and geography of the disposal site. Even concentrating the radioactive fission products from liquid waste into solid form, leachable or not, would be a major step forward. Once they are robbed of the mobility of liquid flow, the fission products are far less hazardous.

In theory, many possible methods of fixation are available. The investigations at Oak Ridge have been limited to mixing the waste with clay and certain common fluxes and heating the mixture to form a brick or sinter. In particular, these investigations have been concerned with the possibility of using heat from the decay of the fission products in the waste to form a sinter. This process would be self-contained and, therefore, safer. Two problems are involved: First, the development of a mix which will form a satisfactory solid at a temperature low enough to be reached by fission product decay-heat; and second, the design and engineering developments which will convert a laboratory procedure into a routine operation.

The first sintering experiments at Oak Ridge were centered on the use of Conasauga shale, a local geologic formation, literally as "cheap as dirt." Preliminary tests with tracer quantities of radioactivity were very encouraging. Later tests with actual high-level waste, however, showed that some of the sintered cakes were far more leachable than we had been led to expect from our early work. In particular, mix 15, the best of some 200 waste-clay mixes, would not retain cesium when the sintered mix was put in water. This led to a period of considerable reevaluation, particularly as to the practical need for making the solid resistant to leaching. Should we plan to store the solid where it would remain dry indefinitely and put all our effort into obtaining maximum physical strength and mechanical stability? Or should we design a sinter so unleachable that it could be stored safely in any convenient location? For the present at least, we have decided to continue working toward our original goal of a leach-resistant solid.

The leachability of a sinter depends on several properties, including surface area and solubility. A porous or powdery sinter exposes more surface area to be attacked by water than one which is dense and impermeable. The differences in the physical properties of a sinter depend on composition, particle size, degree of mixing, rate of heating, and so on. The art of baking "mud pies" so that they will always be dense and impermeable is one that we are still learning.

A more fundamental factor in leachability is the way in which the various radioactive elements are held in the solid material. The sinters are composed largely of very fine grained crystals, invisible by optical means but easily identi-

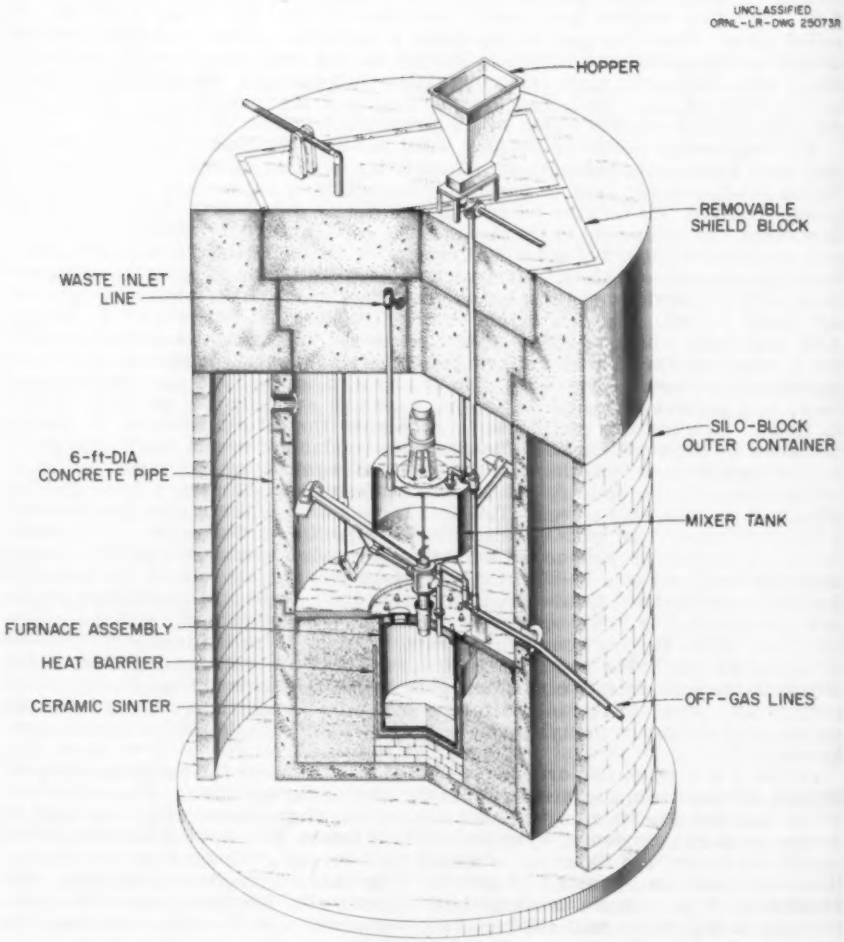
fied by X-ray diffraction analysis. Like all crystals, they consist of a lattice, or network; the snowflake is a common example of a type of crystal structure. We find that some of the radioactive elements are held mechanically in the open ings of the crystal lattice, from which position they are easily removed. Other elements, however, are actually built into the crystal lattice itself and become part of it; these elements are not leached out easily. It is probably impossible to find a mixture which will build all of the fission products into the crystal lattice. For this reason, mechanical retention within the sinter must also be relied upon. Operation limitations make it desirable to form the fired sinter at as low a temperature as possible. We are having some success with our search for a mix which will yield better crystals. For example, the addition of boron in the form of boric acid, has greatly improved the retention of cesium, and we are studying changes which will improve the retention of strontium.

The engineering design of the pilot plant in which we hope to make a practical test with substantial amounts of radioactivity is based on our desire to use the fission product decay-heat to fire the sinter. This is not to save the cost of the power required, but because we feel the job can be done better this way. In the first place, the equipment is simpler and the combination of unbroken insulation and shielding around the sides and bottom of the sintering furnace provides a desirable element of safety. Heaters immersed in the sinter must have connections with the outside and these connections, in addition to the heaters themselves, are likely to fail. The heat from radioactive decay is not subject to failure. Also, this heat, which would be evenly generated within the waste-clay mixture itself, would produce a more uniform product without overheating some parts and underheating others. The cake, after it starts to harden, is a poor conductor of heat; and heaters imbedded in the cake tend to overheat the material nearest to them. Overheating can be serious because the off-gas problem is greatly increased by the volatilization of some fission products if even a small part of the mix is heated above the minimum temperature required for sintering.

The design of the pilot plant for self-sintering encountered a heat problem which we feel we have solved. The heat losses per unit volume are greater with a small furnace than with a large one because the surface area of the small furnace is large in proportion to its volume. There is a minimum size below which sintering temperatures (~850° C.) cannot be reached because of the practical limitations of thermal insulation. The upper limit on size is determined by the hazards involved in handling large volumes of radioactive waste at our particular plant site. The compromise has been to build a small furnace surrounded by a heat barrier. The heat barrier is a blanket of exterior heating coils which will keep the furnace wall almost as hot as the sinter inside of the furnace, and so prevent heat loss. This "adiabatic" furnace will permit the waste to heat and sinter itself exactly as though we had a unit several times this size without a heat barrier.

Figure 1 is a schematic drawing of this plant. It shows the outer containment vessels, mixing tank, furnace, heat barrier and radiation shield. For safety the mixer and furnace unit requires an off-gas cleaning system with a variety of devices such as a condenser, caustic scrubbing tower, jet mixer with recirculating caustic absorber, and filters so arranged that we can evaluate several combinations, and can sample at many points. The characterization of off-gases and evaluation of this system are important objectives in this experiment. In operation the liquid waste and solid materials are added to the mixer through the shield block and, after mixing, are dumped into the furnace. The sinter, once formed, can be removed either for testing or disposal, and the same plant used to fire more sinters. The pilot plant is now 90 percent complete, and we are awaiting the completion of our laboratory work on sinter development to make the initial run.

We cannot at this time foresee all of the engineering developments which might lead to "self-sintering" as a feasible and economical method of ultimate disposal. It is doubtful whether it will ever become a production-line operation along the lines suggested by our laboratory and pilot plant equipment. It is reasonable, however, to consider the application of self-sintering as an adjunct to tank storage. The problem of mixing radioactive wastes in the tank with inert additives to produce the ultimate sinter could be accomplished by a remotely operated, closed-loop arrangement of pumps and mixers. The wastes would be pumped out of the tank and the waste-clay slurry pumped back in. Repeated recirculation through such a system should provide adequate mixing. Following this, the mixture would self-sinter at the required temperature. Certainly, the self-



Experimental Facility for High-Level Self-Sintering.

FIGURE 1.

heating cycle would be more easily accomplished in a large, underground storage tank than it has been in the small furnaces used in our laboratory.

Undoubtedly we will continue to store these high-level liquid wastes for some time to come. For the present, tank storage is the safest and possibly the cheapest method of containing the wastes. However, we do not regard containment of radioactive liquids in tanks as an answer to the problem of permanent disposal. For reasons of safety, it would be better to self-concentrate the liquid to a solid residue in the bottom of the tank. Even this residue would be subject to decomposition and possibly too soluble to store there indefinitely. When these factors are considered, it would appear even better to develop a method by which wastes in the storage tank would self-sinter to produce an insoluble ceramic in which the long-lived fission products are permanently fixed. Fixation and storage in tanks provides safe containment during the preliminary cooling period and has the added advantage of stability and, hence, greater safety for permanent storage in situ.

Representative HOLIFIELD. Our next speaker is Mr. R. L. Doan of Phillips Petroleum Co., on the same subject.

STATEMENT OF R. L. DOAN,[3] PHILLIPS PETROLEUM CO.

Mr. DOAN. Gentlemen, I would like to make a statement on behalf of Phillips Petroleum Co. as operator of the Idaho Chemical Processing Plant relative to the waste disposal research and development program on the fixation and ultimate disposal of high-level wastes at the Idaho Chemical Processing Plant.

The ultimate disposal of high-level wastes from the processing of spent-fuel elements has been generally recognized as one of the most important problems in the atomic energy field. The storage of radioactive wastes as liquids has always been considered a temporary and not a final solution. Ultimate long-term safety is not assured by this method because of possibilities of tank rupture due to corrosion, fabrication defects, or major ground disturbances which would release radioactivity. Such storage is quite expensive as indicated in previous reports to this committee.

For example, as I pointed out yesterday, the investment cost of underground storage tanks at the Idaho Chemical Processing Plant is about $2.63 per gallon for aluminum alloy wastes and $4.90 per gallon for zirconium-alloy wastes.

The search for more effective and more permanent disposal methods is being conducted at many institutions. The best solution will undoubtedly involve the reduction of the wastes to solid form for storage.

Some advantages are—

1. Since solids can be made in forms similar to earth materials, they should be retainable in a storage location indefinitely. The loss to ground of contained fission products even without additional fixation should be many orders of magnitude lower than that of fission products in solution.

2. In the event of an earthquake solids are relatively immobile.

3. Since water is removed in converting to solid form, storage volumes are markedly reduced, except as inert materials are added.

4. Containment structures can be simplified since solids are less mobile and less corrosive than liquids.

[3] See biographical sketch, p. 598.

5. Earth-like solids appropriate as storage media, though poorer conductors of heat than liquids, may safely be allowed to reach much higher temperatures in storage.

Fluidized bed calcination: In keeping with the responsibility of operating the Idaho Chemical Processing Plant, Phillips Petroleum Co. has explored the field of solids preparation and disposal, has studied the approaches which others have tried, and has picked one promising attack for detailed study. The aim has been to develop an economical and mechanically simple process which will provide complete containment, or, at least, a significant reduction of the possibility of fission products escaping to the environment. Laboratory and pilot plant work during the past 2 years has developed the process sufficiently to warrant the construction of a small demonstration plant. This work has been confined mostly to handling aluminum nitrate wastes which constitute the bulk of the ICPP wastes at this time.

There are approximately 750,000 gallons of these aluminum nitrate wastes stored in underground tanks at the chemical-processing plant as a result of process operations to date.

Briefly, the process involves flashing the liquids directly to solids on a high-temperature bed of previously produced solid particles. The advantages of such fluidized bed calcining operation, as compared to other methods for the production of solids, are—

The absence of moving parts in the calcining device and in most process steps;

Its easy applicability to continuous processing;

The efficiency with which heat required for decomposition is transferred;

The ease with which the product may be transferred to storage; and

The utilization of the aluminum oxide as the fission product retaining medium.

The process steps consists of fluid bed calcination, treatment of the offgas to remove volatile fission products and entrained radioactive dust, and storage of the solids in underground concrete vaults. These steps are described in somewhat more detail in the following paragraphs.

The calciner consists of a cylindrical vessel containing a bed of granular aluminum oxide. The bed is supported on a perforated plate which serves as a gas distributor. Preheated air is passed upward through the plate to fluidize the bed, the entire dense mass behaving much like a vigorously boiling liquid. The resulting high state of agitation ensures complete mixing of the bed and provides excellent conditions for the transfer of the heat required to evaporate the water present and to decompose the nitrate salt. Additional heat is supplied through the vessel wall or through heat-transfer tubes within the bed itself. The bed is maintained at a temperature within the range of 750° to 1,000° F.

The waste solution is injected directly into the bed through spray nozzles. The liquid droplets, upon coming into contact with the hot solid particles, are dried and their contained solids are decomposed to oxides. The solid product is deposited upon the particles in the bed and these particles are continuously withdrawn from the bottom or from an overflow pipe.

The calcined product, consisting of sand-like granules of alumina containing the fission products, is transported by pneumatic means to the storage vessels. Because of the refractory nature of alumina, it can be allowed to reach relatively high temperatures in storage as a result of the heat liberated by fission product decay.

The gases leaving the calciner consist of air, steam, and oxides of nitrogen. Entrained dust particles, which will be highly radioactive, are removed by scrubbers, cyclones, filters, silica gel adsorbers, et cetera, before release of the gas to the atmosphere.

Water-vapor and nitric-acid fumes are largely discharged to the atmosphere. At other sites, where a particular economic or environmental situation would justify such a process step, the nitric acid could be recovered for reuse or neutralized for normal disposal.

Present status: the present status of the development program can be summarized as follows:

1. The operable feasibility of the calciner has been established in pilot-plant equipment, working with nonradioactive simulated waste solutions. A scale-up model is being built to investigate more optimum conditions of operation.

2. Ruthenium appears to be the only significantly volatile fission product at calcining temperatures up to 1,000° F. Small-scale laboratory experiments show that this element should be removed adequately from the gas by scrubbing with dilute acid and/or adsorption on silica gel beds.

3. Unsolved problems still exist in the removal and retention of very fine radioactive dust carried in the offgas stream. It has not been proven conclusively that this dust can safely be released to the atmosphere.

Studies have been conducted on porous metal filters, cyclones, scrubbers, electrostatic precipitators, and absolute types of filters. The data obtained on these devices indicate that all but the smallest particles can be removed with excellent efficiency. No sampling and analytical methods exist which will determine the quantity or particle size distribution of very fine dust from "cold" material with sufficient accuracy to evaluate the radioactive release from the calcining plant to the atmosphere.

Work is continuing on the evaluation of gas cleaning equipment, but the final answers, including operating efficiencies of scrubbers, cyclones, et cetera, can only be obtained by the use of hot material and by using equipment of engineering size.

It should be mentioned that the production of these fine particles is encountered more or less in all gas-generating processes for converting liquids to solids, but is somewhat more acute in a fluidized bed process.

The effective removal of dust from vented gases has been accomplished in many industrial operations, but its retention for the present purpose must be orders of magnitude better due to the radioactivity which is present.

4. In the storage of the solid wastes there is a possible problem with respect to the removal of the heat generated by the radioactive decay of the fission products. A cubic foot of the fission product-bearing alumina produced by the calciner, if secured from highly burned up reactor fuel and after the usual 4- to 6-month decay period, is calcu-

lated to generate heat at a rate of about 800 watts, which is about that of a household toaster. Since alumina is a good insulator, this heat is not easily conducted from the center of the mass to the outer surfaces. Hence, inside temperatures can rise to very high values. As the shorter lived fission products decay, the rate of heat generation decreases until after 5 years it is only about 1 percent of the above figure. At that time the fission products cesium and strontium are the principal sources of heat.

Experiments are being conducted in the ICPP hot cell to produce about a cubic foot of full activity level wastes for direct studies of heat generation and removal.

Experiments have demonstrated that there is a good possibility of removing practically all of the cesium, about three-fourths of the strontium, and a few percent of other fission products by leaching the alumina with hot water or dilute acids. Thus there is a possibility of alleviating the heat problem for long-term storage. Further, such leach solutions can be treated to concentrate these fission products for sale as radioactive sources.

Future program: As mentioned earlier, development has reached the stage where larger facilities are required and construction of a semiworks plant at the Idaho Chemical processing plant is underway. The objectives of this plant are—

1. Resolution of problems which can only be demonstrated at full activity level and on equipment of engineering size.
2. Provision of quantities of solid wastes for study of the storage problems, further development work on fixation of fission products, and other studies.
3. Demonstration of the process and study of the operational behavior of the plant.
4. Collection of engineering data for design of full-scale plants.

Engineering design is essentially complete and construction is in progress for a 60-gallon-per-hour facility. The 4-foot diameter calciner will be operated at about 750° to 1000° F. The simple, but highly efficient off-gas components will include high efficiency cyclones, wet scrubbers, adsorber filters packed with silica gel, and positive AEC-type filters. Provision is being made for installation of additional or alternate gas-treating components should the initial system not prove sufficient.

Calcined alumina will be transferred by an air stream to the solid storage vessels. Sufficient storage volume will be provided initially to store about 1 year's production of solids. Since the behavior of the radio-active solids when stored as a powder in large mass is inadequately known, the central temperature of the mass of solids will be limited to that at which calcination is conducted. Thus fission products which are not volatilized in the calciner should not volatize in storage. The solids will be contained in narrow vertical annular chambers with intervening passages for cooling by forced air. The design is based on storage of wastes 2 years after reactor discharge since wastes of this age do not generate as much heat and will be available for study, and since it is possible to store younger wastes as liquids in the tanks available at the ICPP.

For storage of further quantities of alumina produced by the calciner, advantage will be taken of information developed in the

plant to permit simplification in design and reduction in cost. A special experimental storage facility is planned in which central temperatures in the alumina can be allowed to rise under control to very high levels in order to determine the feasibility of uncooled storage.

No equipment is being planned in the demonstration plant to permit leaching and recovery of the fission products from the alumina. Development activities on this phase of the program will be carried out in the general ICPP hot pilot plant facilities. The most effective equipment can be added later to the demonstration plant.

The development and demonstration of this fluidized bed calcination process represents an important advance in the solution of the waste-disposal problem. In principle, this same process can be applied to types of wastes other than nitrate wastes derived from aluminum-type fuels. Exploratory tests on simulated wastes from processing zirconium and stainless steel fuels indicate that these fluoride and sulfate containing wastes are readily calcined to granular solids. As soon as time and manpower are available, investigations of these wastes will be undertaken.

In connection with the operation of the processing plant in Idaho, Phillips Petroleum Co. has been investigating, and will continue to investigate, ways and means of reducing the cost of present liquid-storage methods. Programs which are under way include reduction of the quantity of liquid wastes by changes in the fuel-processing system, separation of the bulk quantities of inactive components, such as salts of aluminum, zirconium, and iron, from the relatively small quantity of radioactive constituents, and finally, the neutralization of acidic wastes to permit storage in cheaper tanks. Presently we use stainless-steel tanks. An effective way of neutralizing the wastes so they can be stored in something cheaper than stainless steel would, of course, be an advantage, if you have to stick with liquid-storage methods which presently we do not believe to be the case.

Representative HOLIFIELD. Does this experimental work you are doing stem from the $3,200,000 authorization we made last year in the fiscal 1959 budget?

Mr. DOAN. I presume it does. The cost, I believe, of this total facility is of the order of $5 million.

Representative HOLIFIELD. Is this project sufficiently financed?

Mr. DOAN. I believe we are all right.

Representative HOLIFIELD. Do you know if there is an item in the 1960 budget for extension, or not?

Mr. DOAN. I am not sure of that.

What we would like to do is to learn as much as we can with reference to this plant and if it looks feasible then to come in with an item for a larger plant which will take care of all of the aluminum alloy waste.

Representative HOLIFIELD. Are there any further questions?

Representative HOSMER. The process you described has the purpose of turning wastes into solids of some sort?

Mr. DOAN. That is right.

Representative HOSMER. Whereas the previous witnesses were anxious to turn them into solids of a particular sort that are not subject to leaching and so forth?

Mr. DOAN. What we are really trying to do is to take the large bulk of waste, solution waste, presently in underground tanks, just as it is, and turn it by an economical method into solids. This results in two things:

A big reduction in volume and the permissibility of storing this in something less than costly stainless steel tanks, underground concrete vaults, or something like that, where you don't have the corrosion problem, or the dispersal problem, or the leaking problems that you have with liquid wastes.

Representative HOSMER. You do point to a further reduction in volume factor in connection with cesium and strontium by the paragraph in connection with leaching.

Mr. DOAN. Yes.

Representative HOSMER. If you can get those out you have a further reduction factor, but you still have the problem of some kind of container.

Mr. DOAN. Principally you get out the cesium which is one of the chief heat generators, the cesium and strontium, and, of course, also those are two of the most likely radioisotopes which can find commercial use.

We know we can leach them out with water. It may prove practical to do it. It is a question of whether the cost of the plant to do it can be justified on the basis of reducing the heat generation rate and perhaps by a market for the cesium.

Representative HOSMER. Thank you.

Senator DWORSHAK. Mr. Doan, will it be necessary to store or immobilize those wastes permanently, or will it be possible to eliminate the waste materials which may cause contamination?

Mr. DOAN. One will have to store them over a period of several years. If you could get out the cesium and strontium, which are long-lived radioistopes, with half lives of the order of 25 to 30 years, this would be very helpful.

Unfortunately, as Dr. Wolman has pointed out, you never get them out so completely that you can just go and shovel the residue out on the ground some place. You will still have to keep it retained.

Senator DWORSHAK. There will always be a threat from those waste materials so long as they are even underground?

Mr. DOAN. It is a long-term problem. I would say that the hazards are substantially reduced by turning them into solids.

Representative HOLIFIELD. Thank you, Mr. Doan.

Before Mr. Heroy speaks, we have several statements for the record.

(The statements referred to follow:)

THE INTERACTION OF RADIOACTIVE NUCLIDES AND SOILS

An Account of our Present Position with Respect to the Basic Problems Involved

by

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Although this paper is presented in connection with discussions of the fixation and ultimate disposal of the radioactive by-products of the nuclear industry, the authors have felt that they could make their most useful contribution rather by assuming that fixation will never be quite complete and disposal never quite final. We shall, therefore, consider the problems which arise when we must account for the fate of radioactive poisons which have escaped to the environment. It is all too easy to imagine situations in which this type of knowledge will be a necessity, even when the house-keeping of the industry is at its most effective. Even if we totally discount the possibility of a bombing, we cannot rule out the possibility of a nuclear catastrophe. And we must always expect a slow release from control of the undesirable radioactive nuclides. Furthermore, in any disposal operation utilizing the earth directly, a necessary part of the operation will continue to be a constant monitoring of the disposal site and its environs. Any aids to these very expensive operations which may come from studies of the character discussed in this paper would evidently be highly desirable.

In the following we shall arbitrarily limit ourselves to an account of the effects to be expected with water solutions of the nuclides. The fate

of particulate matter in the earth is certainly extremely important, and probably of first importance in connection with radioactive fallout. This aspect of the problem we do not consider, but rather we limit ourselves to the materials resulting from the eventual dissolution of solid particles.

We consider first general types of soils, their distribution, and the minerals contained in them which actively interact with dissolved material. An account is then given of the types of chemical binding between these minerals and dissolved radioactive nuclides. We are led to a survey of the more purely scientific problems which arise when one tries to give a quantitative discussion of the various types of sorption observed. The present state of our knowledge is made evident by a review and discussion of some of the published work in the field. We then pass to more complex problems: Those which arise in attempts to predict the motion of radioactive nuclides in the soil. In conclusion we try to indicate profitable directions in which scientific work should proceed in order to buttress our position in the duty of safeguarding the people of the United States from at least a few of the highly undesirable effects of our rapid technical progress.

I Distribution and Characteristics of Soils

Soils are natural bodies, exceedingly diverse in character. Though classification schemes which include breakdown into detailed properties have been devised, we can look only at the generalized description of soils which enables division into a few major groups. Simonson[1/] has described in a very general manner the distribution of the world's soils into six broad belts, those in each belt having somewhat similar characteristics. Soils in a given belt are further subdivided into great soil groups, of which there are about forty in the United States.

Soil groups bear rather exotic names, largely of Russian origin. Podzolic soils occur extensively in humid regions between 35 and 70 degrees north latitude, and to a lesser extent in comparable regions south of the equator. In the United States, these are the soils of the Northeast and of the humid-region mountains. They are strongly weathered, acid, low in bases and in organic matter. Latosolic soils occur primarily in wet tropical and sub-tropical areas. Weathering to great depths and prevalence of iron and aluminum oxides are prominent features. The red and yellow soils of the Southeastern United States are latosolic in character. Chernozemic soils occur mainly in temperate regions with rainfall between 15 and 30 inches annually, though some are found in the tropics. They are moderately weathered and are of high clay content and base status. Free $CaCO_3$ generally occurs in deeper layers. The prairies of the central United States fall into this category. Desertic soils occur in truly arid parts of the world. Weathering has been slight, and soluble salts, free carbonates, or both, are prevalent. Tundra soils exist in the cold-storage part of the world, and are frozen for much of the year. They consist, for the most part, of slightly weathered rock fragments. Soils of Mountains are a last group. They are stony, generally shallow, and there are inclusions of other soil groups.

Not contained in the general classification indicated above are the locally important Coastal Plain sediments on the Atlantic and Gulf Coasts. This band of soils, extending inland as much as a hundred miles, generally has sandy surface soils underlain by material varying in texture from sand to clay.

Soils containing more than 20 per cent organic matter are termed <u>organic</u>; those with less are <u>mineral</u>. Organic soils such as peats and mucks occur

in local areas, usually of poor internal drainage, and may contain as much as 95 per cent carbonaceous material. At least 95 per cent of the mass of most upland soils is mineral. Oxygen forms the framework for soil minerals and it, hydrogen, silicon, aluminum and iron are the most abundant elements in mineral soils. Except for calcareous soils, which may contain as much as 75 per cent calcium carbonate, and some latosols high in TiO_2, oxides of silicon, aluminum and iron make up at least 90 per cent of the mineral portion. Silicon, aluminum and iron exist in soils in primary, clay and oxide minerals.

The chemical composition in a given soil varies with depth. Because of accumulations of clay and oxide minerals in deeper layers, Al_2O_3 and Fe_2O_3 contents usually are high in subsurface layers, while that of SiO_2 is lower. Organic matter content decreases rapidly with depth. Soil contents of stable Sr, Cs and rare earths usually are very small. On the average, there are some three atoms of Sr per 1000 atoms of Ca.

Most of the common rock-forming minerals in soils occur in sand and silt fractions. As a result of weathering, as well as of parent rock differences, their proportions vary tremendously from soil to soil. Quartz predominates in the sand and silt of humid-region soils, having accumulated at the expense of less-resistant pyroboles, micas and felspars. Average percentages of quartz, felspars and micas in the sand fractions of humid-region soils are 90, 4 and 1.5, respectively. In subhumid and arid region soils, contents of non-quartz minerals in sand-size fractions are larger, averaging 20 and 37 per cent.

Clay and sesquioxide minerals make up the bulk of the clay-size fraction of soils, though others may be present. The proportions of various mineral species vary from soil to soil and within a particular profile. The mineral compositions of clay fractions can be generalized through reference to a

listing of minerals in the order of increasing resistance to alteration. Generally, minerals near the top of the list (taken, with some modification, from Jackson[2]) predominate in clays of slightly weathered soils, such as those in the Chernozemic and Desertic belts. Those near the bottom occur in soils which have been exposed to drastic weathering, as in the case of latosols.

	Type minerals	Type soils in which dominant
Increasing Stability	Felspars	Glacial soils of Southern Canada
	Muscovite-Illite	Subhumid and arid soils, glacial soils of temperate regions
	Vermiculite-interstratified clays	Subhumid and arid soils (from weathering of micas), R-Y Podzolic
	Montmorillonite-Beidellite	Volcanic ash, limestone, basic rock soils. Weathering of micas in temperate regions
	Kaolinite-Halloysite	R-Y Podzolic, some Latosols
	Gibbsite-Allophane	R-Y Podzols, Latosols
	Hematite-Goethite	5-30% in R-Y Podzolic, 30-75% in Latosols
	Anatase	Up to 35% in some Latosols

In zonal soils of humid-cool to subhumid-temperate areas, illite is the predominant mineral. Mixtures of kaolin-vermiculite-interstratified minerals occur in humid-temperate regions. Kaolins with admixtures of smaller amounts of gibbsite and hematite predominate in warm-humid regions. Under tropical conditions, the proportions of sesquioxide minerals are high and in extreme cases only these minerals remain. Arid, basic conditions favor the formation of montmorillonoids. These also occur where soil development is retarded, as by imperfect drainage.

The soil characteristics in which we are most interested, for our present purposes, are those which deal with the sorption of cations and anions.

In clay minerals, as in micas, there is extensive proxying of ions in octahedral and tetrahedral lattice positions. The resulting unbalance of charge requires the presence of cations external to the lattice to maintain electroneutrality, and gives rise to cation exchange capacity. Lattice charges are large for micas and vermiculite (135-270 meq per 100 gm), intermediate for montmorillonoids (80-135 meq per 100 gm) and quite small for kaolins (2-10 meq per 100 gm). In clay mineral micas and in vermiculite, much of the lattice charge is balanced by cations which occur between closely adjacent fixed lattice layers. Inter-layer potassium ions in micas are not displaced under ordinary conditions, and are not counted with the exchangeable cations, though they are bound by the same kinds of forces. The cation exchange capacities of mica clays vary between 20 and 40 meq per 100 gm.

The common interlayer ions of vermiculite are calcium and magnesium, though vermiculite-like minerals with inter-layer aluminum appear to exist in many soils. Rates of ion exchange are low for such minerals, particularly when ammonium or potassium is one of the exchanging ions, and the cation exchange capacity is not well defined. On the other hand, cations balancing the isomorphous substitution charges of montmorillonoids are almost completely exchangeable, and there are excellent correlations between the exchange capacities of these minerals and their chemical compositions. Lattice substitution also appears to contribute to the cation exchange capacity of kaolins, though the charge here is small.

Lattice terminations can result in further development of negative charge, particularly when the pH is above 6. Protons may ionize from SiOH groups

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around the edges of clay particles and edge aluminum ions may adsorb OH^- or may shift coordination from 6 to 4 as pH is raised. Maximum development of edge charge occurs at around pH 10. The magnitude of the negative charge which can develop on lattice terminals depends on edge area. In meq per 100 gm, it is about 20 for montmorillonite, 10 for illite, and 2-10 for kaolin, depending on particle size and crystal form.

Allophanes have cation exchange capacities (pH 7) of from 60-120 meq per 100 gm. Probably this results largely from coordination shifts involving aluminum.

The organic matter of soils consists of a poorly-defined conglomeration of carbonaceous material, but is known to contain carboxyl, phenol, enol and imide groups. Ionization of protons from these results in negatively charged particles, the charge increasing with the extent of ionization. The apparent ionization constant for COOH groups of soil organic matter is about 10^{-5}. Other acid groups ionize appreciably only at pH's above 7. The carboxyl content of soil organic matter varies between about 150 and 275 meq per 100 gm. Organic matter can bind as much as 400 meq of cations per 100 gm at high pH.

Most soils contain two or more clay minerals, with organic matter admixed at least in the layers near the surface. Because clays, and organic matter in particular, correspond to a series of weak acids of varying strength, it is useless to attempt more than a formal definition of cation exchange capacity, since it generally depends on the environment. Perhaps the most realistic view, particularly in acid soils, is that the effective cation exchange capacity equals the sum of the meq of exchangeable cations as displaced by exhaustive leaching with a salt solution. The convention usually employed, that of exchange capacity as the quantity of NH_4^+ sorbed from neutral 1N NH_4OAc, is quite artificial.

Some rather qualitative predictions concerning the exchange capacities of soils can be made from a knowledge of the clay and organic matter contents, and from the kinds of clay minerals present and their capacities, as deduced from material presented earlier. Thus the sandy soils of the Atlantic Coastal Plain have capacities on the order of 2-20 meq per 100 gm, depending largely on organic matter content. Those of the red clays of the Southeastern Piedmont are between about 4 and 10. The Chernozem and similar soils, because of the prevalence of montmorillonite-like clays, often possess exchange capacities as high as 60 meq per 100 gm.

The exchangeable cations are those balacning the negative charges on soil particles. To a first approximation, exchangeable metal cations can be regarded as neutralizing isomorphous substitution charges and those weakly acidic groups which are ionized under the prevailing conditions. Calcium, Mg, K, Na and Al are the most abundant, the last occurring only in acid soils.

The literature of soils is replete with confusing (and confused) statements about exchangeable hydrogen in soils. In reality there is no such thing. While it is possible and even probable that H which is covalently bonded to weak acid spots on clay minerals, and it is certain that H on carboxyl and other groups on organic matter, can ionize and create exchange spots at high pH, in acid mineral soils it is hydrolyzable trivalent Al, not H, which is the exchangeable ion responsible for the observed acidity.

Less weathered soils (because of age, low rainfall, temperate or cold climate) have as exchangeable cations largely calcium and magnesium. Highly weathered soils, except when derived from basic parent material, have large proportions of their permanent latticecharges (20 to 95 per cent under native conditions) countered by exchangeable aluminum. Some soils of dry areas contain large amounts of exchangeable sodium.

The common exchangeable cations differ in their affinity for ion exchange spots on soil clays and organic colloids.

For permanent charges: $Al > Ca \geq Mg > K > Na \geq H$

For weak-acid charges of clays: $H > Ca \geq Mg > K > Na$

For carboxyl groups of organic matter: $H \gg Ca \geq Mg \gg K > Na$

While ion exchange should refer only to the association of ions with solid surfaces in such a way that they can be replaced stoichiometrically by other ions of the same sign of charge, "anion exchange" in soils has a different connotation. In fact, it means different things to different people. Anion exchange does occur in many soils, but often is confounded with other reactions which also result in the lowering of solution concentrations of anions.

Clay and oxide minerals in soils can take up anions from solution and perhaps can develop positive charges on certain exposed surfaces. This might be through proton acceptance, hydroxyl ionization, or perhaps other effects. The apparent positive charges of soil particles increase as the pH is lowered, and in the case of some latosols, high in iron oxide, may exceed negative charge at pH's below about 4. All clay minerals, however, as well as temperate region soils, possess net negative charges at all pH's.

Halide, nitrate and sulfate ions, as well as phosphate, fluoride and carboxylic acid anions, are sorbed by at least some soils, and appear to be mutually replaceable. Capacity for this kind of anion sorption varies from near zero at neutrality for all minerals to as much as 1 meq per 100 gm for kaolins, 20 meq per 100 gm for kaolin-iron and aluminum oxide combinations in latosols. Montmorillonoids and other clays with large lattice charges do not sorb small anions via this mechanism unless salt contents are very high.

Halides and similar anions are negatively adsorbed by such clays under most circumstances.

Certain anions, such as phosphate and fluoride, coordinate strongly with ferric iron and aluminum and are sorbed by soils through another mechanism. Phosphate ions, for example, can react with clay and oxide minerals in soils to form basic iron and aluminum phosphates similar to strengite, variscite and palmerite. Phosphate is displaced from such minerals by hydroxyl, fluoride or other ions which coordinate strongly with ferric iron and aluminum. Such reactions are viewed as involving solution-precipitation rather than anion exchange.

Soils with pH's smaller than 7 are termed acid. Acid pH's usually are due to the presence of exchangeable hydrogen and aluminum ions, the former bonding to weakly acidic exchange spots, the latter to permanent lattice charges. Clays with electrostatically bonded exchangeable hydrogen are unstable, and decompose to yield silicic acid and sufficient Al, Mg or other lattice ions to counter the exchange spots occupied initially by H-ions.

The pH's of acid soils vary between 3 and 7, reaction depending on the ion saturation and the soluble electrolyte content. In the absence of excessive amounts of soluble salts or acids, pH's less than 4 are rare.

Montmorillonoids, vermiculites and illites are similar in their neutralization behavior. Permanent lattice charge accounts for the bulk of their exchange capacity and Al for their acidity. Buffer curves for kaolins have two sections, one indicating displacement of exchangeable Al by Ca, the other reflecting the development of weak-acid charge. Below pH 7, organic colloids behave as weak acids with pKa's of around 5. In the absence of organic colloids and with Ca and Mg as exchangeable ions, pH 6 corresponds closely to 100 per cent base saturation of permanent charge.

Soils containing accumulations of calcium (and magnesium) carbonate are referred to as calcareous. Carbonates may occur throughout the profile or may be concentrated in certain layers, their distributions reflecting the nature of the parent material and the weathering regime. Percentages of carbonates vary from zero to 70.

Calcareous soils are saturated largely with calcium and magnesium. In contrast, with acid soils, pH and concentrations of ions in the soil solution are controlled not by ion exchange equilibria, but by the calcium carbonate-CO_2-H_2O system.

Equilibria in this system can be described by:

$$pH - 1/2\ pCa = 4.85 - 1/2 \log Pco_2.$$

Pco_2 is the partial pressure of carbon dioxide in the atmosphere in contact with a $CaCO_3$-water system, pCa is the negative logarithm of the molar activity of calcium in the soil solution.

Since the CO_2 content of the soil air contents vary from 0.003 per cent, the content in the atmosphere, to nearly 20 per cent, calcium solubility and the reaction of calcareous soils can vary widely.

Soils of dry areas often contain sufficiently large amounts of soluble salts, exchangeable sodium, or both, to have peculiar chemical and physical properties. Such soils are referred to as salted.

The U. S. Salinity Laboratory classification of salted soils is based on the electrical conductivity of a saturation extract rather than on salt percentage per se. Soils yielding saturation extracts with specific conductivities greater than 4 millimhos are saline. The concentration of a solution of this conductivity varies with the nature of the salt, averaging 45 meq per liter for the electrolytes commonly found in salted soils. The

lower limiting salt percentages of saline soils vary from around 0.1 for coarse-textured soils to 0.25 for heavier ones.

The second classification criterion for salted soils is their percentage sodium saturation. If this is above 15, the soil is alkali.

The salts in salted soils largely are chlorides and sulfates of calcium, magnesium and sodium. The proportions of the cations in solution are controlled by ion exchange equilibria.

Saline-alkali soils are changed into alkali soils when salts are leached away. Similarly, normal or saline soils can be converted to alkali soils through the use of irrigation water containing large amounts of sodium.

Most of the preceding has dealt with soil properties relating specifically to their character as ion exchangers. Brief additional mention should be made, however, of their water transmission properties. In this, we are concerned especially with rates of water flow which may be encountered, so that we can reach some conclusions regarding the approach to equilibrium of reactions involving solutes and soil particles.

Soils are porous media, but differ in total porosity and particularly in pore size distribution, depending on the sizes of primary particles and the ways in which they are arranged. Vertical inhomogeneity is the rule. The flow of fluids through soils follows Darcy's Law, which says that flow rate per unit area is proportional directly to the hydraulic head and inversely to the distance traversed. The "hydraulic conductivity" is the proportionality constant in Darcy's equation, and is the flow rate for unit length and cross-sectional area for unit head. Mason, et al.[2] have summarized the hydraulic conductivities of soils from all over the United States, determinations from around 10,000 samples being considered. Averages

for surface layers range from 0.13 to 9.27 inches per hour, with a mean of 2.99. Smaller hydraulic conductivities generally are found for subsoils. Values range from 0.12 to 3.63 inches per hour, with a mean of 0.91.

Since the above flow rates are for a one-inch column and since flow is inversely proportional to column length, it is quite apparent that water movement through soils is a slow process, and that non-equilibrium effects during many leaching processes may not complicate matters in too serious a fashion.

II The Binding of Radioactive Nuclides in Soils

The great variety of relatively long-lived radioactive nuclides in the fission wastes is, unfortunately, such that we can expect no significant simplification of our problem through a limitation in the number of chemical elements to be considered. Even if this were not so, the composition of soil waters is sufficiently varied and so great a variety of inactive salts may be expected to accompany the radioactive elements, that these may at some time be found in competition for the soil minerals in nearly every conceivable inorganic chemical environment. We thus may as well attack the problem of adsorption on soils with little or no regard for the practical ends of the work. Indeed, in such complex survey work it is probable that useful results will be more quickly found through systematic basic research than through attempts to solve specific problems before these actually arise.

It is, however, easily possible to summarize the types of sorption chemistry we may expect to encounter in terms of important long-lived nuclides. We will include nearly the whole field of relevant sorption phenomena in considering the behavior of the five elements Cs, Sr, Ce, Ru, Pu.

In its sorptive behavior cesium differs but little from potassium and somewhat more from sodium, elements with which it will certainly be found in many practical problems. In these cases we have to deal with singly-charged ions which do not exhibit any very complicated behavior in solution and which quite certainly bind to mineral surfaces simply because of the presence in or near these surfaces of negatively charged sites or perhaps of negative ions loosely bound through other forces. To the list of singly charged cations we must, of course, add the hydrogen ion. Its special properties may be of primary importance when dealing with soils containing considerable organic matter and, consequently, insoluble weak acids.

Much the same sort of remark applies to strontium, which will always be associated with calcium and frequently with magnesium in the soils. The behavior of the dipositive ions is apparently influenced by strong interactions with water. With the more weakly basic ions, particularly magnesium, the effects of hydrolysis may become of importance; again we must take the hydrogen ion into account.

Very complicated sorptive behavior is found with the trivalent elements. Even if it were possible to disregard the tripositive ions resulting directly from fission - they are relatively short-lived - we certainly cannot disregard the interactions of aluminum with the soils. Aluminum is frequently present in fission wastes, and in any case it must always be considered whenever soil minerals are exposed to acid solutions when aluminum appears in solution as a result of mineral decomposition. As we shall see, the presence of aluminum has a very marked effect upon the behavior of other elements in contact with the soils.

The element ruthenium might well serve in one or another form as an example of every type of sorptive behavior. The solution chemistry of this

element is so extraordinarily complex that it appears to be impossible to give any useful account of its sorptive interactions. Fortunately, the radioactive nuclides are of relatively short life. We shall here attempt no further discussion of ruthenium.

It is not now possible for us to give a useful account of the sorptive behavior of plutonium, or, indeed, of the other chemically similar heavy elements. We, therefore, limit our consideration almost entirely to effects to be expected with chemically stable singly-, doubly-, and triply-charged ions as exemplified by cesium, strontium, and the rare earths.

Before turning to detailed results we attempt a classification of the types of sorption to be expected with the various minerals in the soils. It appears reasonable to suppose that we can consider the sorptive agents to be of two main types. These "pure" types, and cases corresponding to "mixtures" of them in various proportions, will serve to describe qualitatively the considerable variety of behavior that is observed. Perhaps the simplest type of sorptive action is that due to ion exchange on fixed charge sites, either positive or negative. The primarily sorbed ions neutralizing these charge sites will generally be nearly in equilibrium with the ions of the surrounding solution. The qualifying adjective "nearly" is necessary in the event the surrounding solution is in motion with respect to the solid material. As we have seen, in very many practical cases the motion of soil water is so slow that it will be quite unnecessary to consider rate effects. In the laboratory, however, many of these sorptive phenomena are most readily investigated in flowing systems, with the use of a chromatographic "column". In such experiments non-equilibrium effects are very apparent, and these rate-dependent effects must be considered in order eventually to arrive at data

referring to equilibrium state. We will return to this point. For the moment we assume that we can obtain true equilibrium data and so can recognize cases of simple ion exchange. The behavior of several of the simpler clay minerals seems to be describable in these terms. Thus, with the montmorillonite clays certainly the most important effects are due to the exchange sorption of cations, although undoubtedly these minerals contain also a small proportion of positive sites and so interact directly with the anions of the solutions. The cation exchange properties of the clays have been much more thoroughly studied than their anion exchange characteristics. Perhaps only with the kaolins are the two effects of about equal importance. Anion exchange on the clays is difficult to study because of the small effects and because it can easily become confused with the second principal type of sorption, which we now consider.

The hydrous metal sesquioxides are important constituents of the soils and are often intimately associated with the clays. When these substances are present in appreciable quantity, as is true with the widely-distributed red clays of the East and Southeast, we have to deal with an essentially different sorptive effect: The sorbent may become invaded by a neutral salt, or better said, it is invaded by a neutral combination of anions and cations in proportions which depend on the difference between the environment within the oxide gel and in the surrounding solution. This effect is exactly similar to the so-called "Donnan invasion" of the particles of a resinous ion-exchanger. The effect has been extensively studied with the resinous exchangers[4]. With the soil minerals, although the effect has long been known, the systems are much more difficult to define, and easily interpretable data on simple systems are hard to find.

It is evident that many possibilities can arise, depending on the proportions in which these types of sorption are mixed in various cases. We proceed to give selected examples as illustrations.

In most instances, when trace amounts of cationic radionuclides of the sort we are particularly concerned with are added to soils, major fractions are sorbed. Though little or no theoretical understanding can be gained from examining the results of such experiments, it is considered of some value to indicate the gross effects of the sorption processes.

Strontium in trace concentration is sorbed in most soils largely, if not entirely, as an exchangeable ion, though the possibility of complexing reactions with organic matter functional groups and perhaps other modes of binding cannot be ruled out. Of course, in soils containing free carbonates, sorption on these, with subsequent solid-phase diffusion into crystals may occur. Cesium and trivalent rare earth ions certainly are sorbed by ion exchange, though other binding mechanisms may be operative as well. Particularly with Cs, we are faced with the knotty problem of unraveling the matter of ion-fixation, since Cs may enter mica-like structures as do K and NH_4. As shown in Barshad's[5] work, ions of the proper size to fit into depressions in the oxygen framework of certain layer silicates can, though bound by the same electrostatic forces operative in exchange sorption, be exchangeable with difficulty, or sometimes not at all.

McHenry[6] studied the sorption of Sr-90 by soils from the Hanford vicinity and from other parts of the Northwestern United States. $SrCl_2$ solutions varying in concentration from 10^{-2} - 10^{-7}M were added to soil samples in amounts to supply Sr from 10^{-4} to 10 times the exchange capacity of the soil and distribution coefficients ($Kd = \frac{\text{Sr/gm soil}}{\text{Sr/ml solution}}$) were determined. As would be expected, Kd varied from large values (70-400) in

dilute (10^{-7}M) $SrCl_2$ solutions to less than 1 when amounts of Sr larger than exchange capacity were added. The author makes the point that in the dilute region, with Sr added <0.1% of exchange capacity, Kd is a constant for a particular soil. No relation was found between Kd and exchange capacity, though the two largest were found for high capacity soils.

When $SrCl_2$ concentration was $3x10^{-7}$M, the addition of KNO_3 to the soil-Sr mixture in amounts up to 10 times that of Sr did not affect Kd appreciably. At the same Sr concentration, Cs, Ba and La chlorides (10 x Sr concentration) reduced Kd about 15 per cent. Sodium salts of nitrate, chloride, oxalate, sulfate and phosphate had little effect on Kd at low ($3x10^{-6}$M) $SrCl_2$ concentrations. When $SrCl_2$ was $3x10^{-4}$M, oxalate and orthophosphate increased Kd tremendously, presumably through precipitation.

Addition of HCl to the soil-$SrCl_2$ mixtures to adjust pH downward, drastically reduced the distribution coefficient (and the amount of Sr sorbed by the soil), regardless of the $SrCl_2$ concentration. In part this must be due to the tremendous increases in electrolyte concentration accompanying the acidification of a calcareous soil. Other factors undoubtedly include the destruction of pH-sensitive exchange sites and the introduction of exchangeable Al upon acidification.

McHenry also studied the desorption of Sr-90 (<1% of capacity) from soils on leaching with water or with electrolyte solutions. Water and 10^{-3}N $NaNO_3$ removed the same amounts of Sr, 140 ml per gm of soil being required to elute one half. About 1/4 as much 10^{-3}M $Ca(NO_3)_2$ and less than 1/15 as much 10^{-3}N $La(NO_3)_3$ or 10^{-2}N HCl were needed to do the same job. Sodium salts of carbonate and orthophosphate were much less effective in replacing soil-adsorbed Sr than were Cl, NO_3 or SO_4 salts.

Rhodes[7] also examined the uptake of a number of radioactive materials (trace levels) by Hanford site soil as affected by pH and by NaCl concentration. Kd was found to be independent of soil-solution ratio and of quantity of added nuclide, and was used as a sorption index.

Kd for Ca (from water) was >200 at pH 3.7 to 10.1. From 4M NaCl, Kd was 18 over the same range of reaction. Kd for Sr varied linearly from ~3 at pH 5 to 120 at pH 10. In 4M NaCl, Kd at pH 10 was only about 8. However, the addition of 10^{-2}M sodium phosphate to the soil-Sr90-NaCl mixture increased Kd to values above those obtained for water alone at all pHs.

Distribution coefficients for other nuclides showed highly individualistic behavior characteristic of their horribly complex solution chemistry. Kd for Pu-239 was very large between pH 2.2 and pH 8.4, falling off on either side of this range. Ru-106, Y-91, Zr-95, Nb-95 and Ce-144 were taken up strongly at pH's between about 6 and 8. Uptake was smaller between pH 8 and 11, increasing again at very alkaline reaction.

Nishita, et al.[8] studied the uptake of Sr-90, Y-91, Ru-106, Cs-137 and Ce-144 by a number of soils and by kaolinite and montmorillonite, "fractionating" the nuclides added to a given soil into "water soluble" (that removed on washing 1 gm with 50 ml H_2O), "exchangeable" (that removed by leaching a water-washed sample with neutral 1N NH_4OAc) and fixed (that not removed as above). Recovery in the several forms was more or less independent of dose (in the trace region). Except for a muck (organic) soil, at least 80 per cent of the Sr-90 was either soluble or exchangeable. With Cs-137, on the other hand, from 40-60 per cent of dose was not recovered by NH_4OAc leaching. Similarly, except for the muck soil, 50-95 per cent of added Y-91, Ce-144, and Ru-106 (added as chlorides in HCl) were "fixed". How much of the apparent fixation was due to the participation of the ions in reactions

other than ion exchange and how much to possible large exchange affinity at trace concentrations is not known.

Shulz, et al.[9] have taken a careful look at the condition (exchangeable or fixed) of trace levels of Sr-85 in twenty-six California soils varying in capacity from 3.5 to 26, in pH from 5.6 to 8.3 and in free $CaCO_3$ from 0 to 13 per cent. Soils were mixed with carrier-free Sr-85Cl_2, dried at either 50 or 110°C for one week to enhance "fixation", and extracted (either batch or leaching) with 1N NH_4OAc or 1N $CaCl_2$. Though removal of Sr-85 was incomplete (1-7 per cent of dose remaining after most efficient extraction), the slope of the removal curves did not reach zero, and the authors feel there is no evidence for the existence of Sr in other than exchangeable form in any of the soils examined. Of interest is their treatment of serial batch extractions of Sr-85 with 1N NH_4OAc. Using a mole fraction exchange equation, they computed the expected removal of Sr for each batch extraction, and compared this with observations. General agreement indicates again that ion exchange equilibria alone were involved and that results for a number of diverse soils can be generalized in terms of this.

A translation of some of the early Russian work on the behavior of fission products in soils[10] shows that the Russians have done many of the same kinds of things done in this country and that they reached much the same conclusions. From batch experiments with podzol, black earth and red earth soils, trace quantities of Sr-89 were strongly sorbed, but could be displaced completely by 1N $CaCl_2$. One N $NaNO_3$ also displaced Sr completely from the red earth, but was not so effective with the other soils.

Cs-137 (trace amounts) was sorbed more completely than was Sr-89. One N $CaCl_2$ affected some displacement of Cs, amounting to about 10 per cent in the podzol and the black earth, 35 per cent in the red earth. One N $NaNO_3$

displaced Cs somewhat more effectively than did Ca. One N KCl was a far more potent exchanger of Cs from the podzol and black earth, removing 59 and 74 per cent, respectively. The amount exchanged from the red soil was no different from that removed by Ca or Na.

Trace amounts of Zr-95 - Nb-95 were sorbed almost completely by the podzol and black earth soils, less by red soil. One N $CaCl_2$ affected virtually no displacement.

The Health Physics Group at ORNL[11/,12/], faced with practical problems relating to waste pit disposal, have studied the holdup of activities present in waste pit liquids when these are passed through columns of Conasauga shale. The waste solutions contain 600 meq Na and 340 meq NO_3 per liter, with lower concentrations of other assorted ions. The pH is around 10. The cesium-137 activity is 0.66 meq per γ_o, corresponding to a concentration of 2.5×10^{-7} M. Also present are much smaller amounts of Ru, Co, Sb, Sr, rare earths, Zr and Nb.

Conasauga shale has an exchange capacity of 22.3 meq per 100 gm. The breakthrough capacity for Cs-137 in the waste mixture is 0.35 millicurie per gm, corresponding to 0.01 meq of Cs held up per 100 gm of shale. With Cs-137 in water (2.4×10^{-7} M), Cs capacity was found in one experiment to be 0.02 meq per 100 gm of shale, or less than 0.1 per cent the exchange capacity. This is a most astonishing finding.

About half of the Cs sorbed from trace concentrations by shale appears to be bound irreversibly, and cannot be displaced by concentrated salt or mineral acid solutions.

As with other soils, the holdup of Cs and Sr is less from strongly acid than from slightly acid or neutral solutions. This is much more pronounced

for Sr. A Conasauga shale column which held up 80 per cent of a Sr-90 dose at pH 5 sorbed only 22 per cent at pH 1.0, 8 per cent at pH 1.5.

When waste solution is passed through Conasauga shale, Sr-90 breakthrough has been found to precede that of Cs-137 slightly (Lacy[12]) or to lag behind (Blanchard, et al.[11]). Lacy found shale columns to hold up around 15 per cent of the Ru in waste solutions, while Blanchard, et al. found Ru, Co and Sb not to be sorbed at all. Rare earths and Y were removed to the extent of about 80 per cent initially, breaking through approximately with Cs-137.

As with clay minerals, Cs has a high affinity for soils. If one calculates an apparent exchange constant (neglecting activity coefficients) for the Cs-Na equilibrium in the shale column, $\sim 1.8 \times 10^3$ is obtained. This is many-fold larger than constants obtained by Krishnamoorthy and Overstreet[13] for montmorillonite ($K_c' = 73$) and Yolo clay ($K_c' = 28$) at macro-concentrations.

From data of Rhodes[7], for the uptake of trace levels of Cs-137 by Hanford site soil, Kd. (Cs/gm soil/Cs/ml solution) was about 16 in 4M NaCl, leading to $K_c' \sim 1.3 \times 10^3$. While there is insufficient data to substantiate the following, it appears either that trace concentrations of Cs may be bound in some non-ion-exchange fashion or that isotherms remain curved in the trace region. It would be most helpful in our presentation, as well as in the far more important matter of adequately describing the behavior of radionuclides in soils, if large amounts of good ion exchange data, extending into the trace region, were available. Unfortunately, this is not so, and we are forced to illustrate points by citing experiments which were conducted at macro-concentrations.

Orcutt, et al.[14] measured Sr-Ca equilibria with 5 oil sands, a Yolo soil (California) and a sample of halloysite. Cation exchange capacities

varied from 0.2 to 25.6 meq per 100 gm. K_c' Sr-Ca (K_c' Sr-Ca = $\frac{qSr \cdot [Ca]}{qCa \cdot [Sr]}$ varied from 1.14 to 1.61, with an average of 1.36. This compares with values of 1.10, 1.35, 1.80 and 2.50 obtained by Krishnamoorthy and Overstreet[13] for montmorillonite, Yolo clay (1.25 by Orcutt, et al.) IR-100 and Dowex-50, respectively and with 1.46, 1.40 and 1.43 obtained by one of us (NTC) for Utah bentonite, White Store soil (montmorillonite-vermiculite) and Davidson soil (kaolin-vermiculite-iron oxide), respectively.

Isotherms determined by eluting Sr-89-labeled columns of Sr-White Store and Davidson soil (NTC) with 10^{-3} M $CaCl_2$, correcting for non-equilibrium with results of isotopic exchange runs, are shown in Fig. 1. Extrapolation into trace regions is uncertain, but nothing particularly odd seems to be going on. One would be tempted to predict that the Ca-Sr isotherm, at least, is linear at low concentrations.

The Al^{3+} ion often occupies large proportions of the exchange sites of acid soils. We are not aware of any published information on Al-Sr exchange equilibria, but one of us (NTC) has obtained Ca-Al isotherms on montmorillonite and on two soils. In one instance the measurements extend almost into the trace region of Ca concentration. Though Ca rather than Sr is the exchanging ion, the rather consistent exchange behavior of the Sr-Ca pair cited earlier makes it appear that Sr would behave in the same way.

Isotherms for montmorillonite and for two soils appear in Fig. 2. Montmorillonite and White Store soil, which owes its capacity largely to montmorillonite, behave much the same, with Al strongly preferred, particularly so when the Ca concentration is small. Davidson, which is a reddish-brown lateritic soil high in iron oxide, holds up Ca in a respectable manner when excess leaching solution is displaced with alcohol. However, on leaching

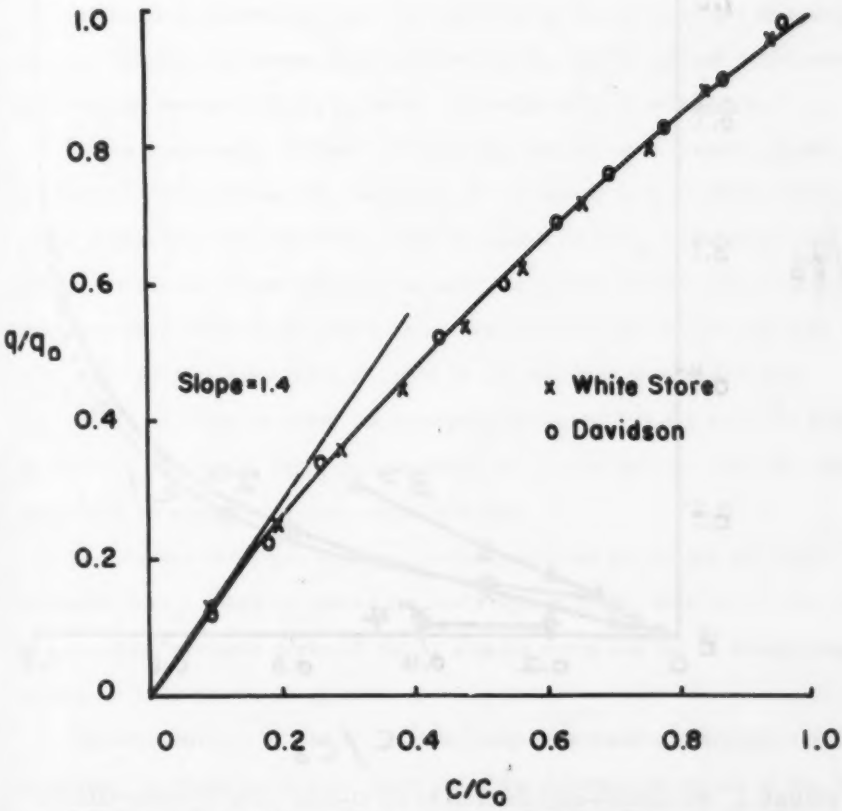

FIGURE 1. The exchange-sorption of Sr by two soils from mixed Sr-Ca chloride solutions with total salt concentration (Co) 10^{-3}M. q/qo is the proportion of the exchange capacity countered by Sr for different reduced concentrations (c/co) of Sr in the equilibrium solution.

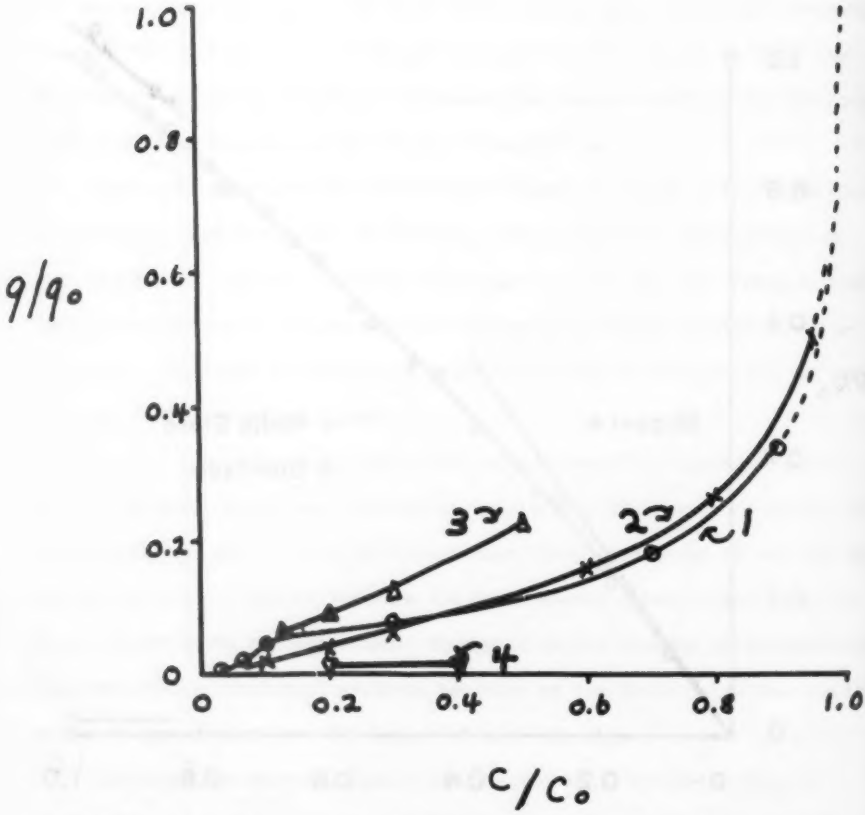

FIGURE 2. The exchange-sorption of Ca by two Al-soils and by Al-montmorillonite from 0.05 N chloride solutions, showing the large affinity of Al for exchange sites. 1 and 4 are for a red soil when washed with alcohol and water, respectively, and show the large readjustment of sorption equilibria during the displacement of "invading" electrolyte. 2 and 3 are for a soil containing montmorillonite-like clay and for montmorillonite itself.

with water, Ca is removed, leaving very small residual Ca content. This must be due to electrolyte sorption from the low-pH leaching solutions, and readjustment of exchange equilibria when solution remaining in the pores is washed out with water.

The large preference of soil (or clay) for Al leads to rather abnormal leaching behavior for trace concentrations of Sr, and to extreme differences depending on whether soil is saturated initially with Al or with Ca.

When a tracer slug of Sr-89 is bled into the top of a Ca-soil column and followed with dilute $CaCl_2$ solution, Sr is eluted in a slightly skewed normal curve with the peak coming when an amount of $CaCl_2$ solution corresponding to 1.1-1.4 times exchange capacity has passed through the column. On the other hand, when Sr-89 tracer is pushed down a column of Al-soil with $CaCl_2$, far smaller quantities of input Ca are required to elute to peak concentration. This is shown for montmorillonite and two soils (10^{-3} M $CaCl_2$) in Fig. 3. Amounts of $CaCl_2$ corresponding to ~10 per cent of capacity were sufficient to elute Sr to peak concentration.

Sr-89 rides the front between Al displaced from the column and the following $CaCl_2$. This is shown (for Ca-45 tracer rather than Sr-89) in Fig. 4, which presents parts of the Al elution curve and the Ca breakthrough curves, as well as the Ca-45 peak.

The odd behavior of red soils, resulting from their electrolyte-sorptive character, is shown in Fig. 5, which shows the relative positions of the Sr-89 peak and the Al elution curve for a Davidson soil column. When tracer Sr was bled into a water-washed column and eluted with 10^{-3} M $CaCl_2$, nothing but water came out of the bottom until 70 ml (~5.4 column volumes) of solution had entered. Then Al and Sr-89 broke through together.

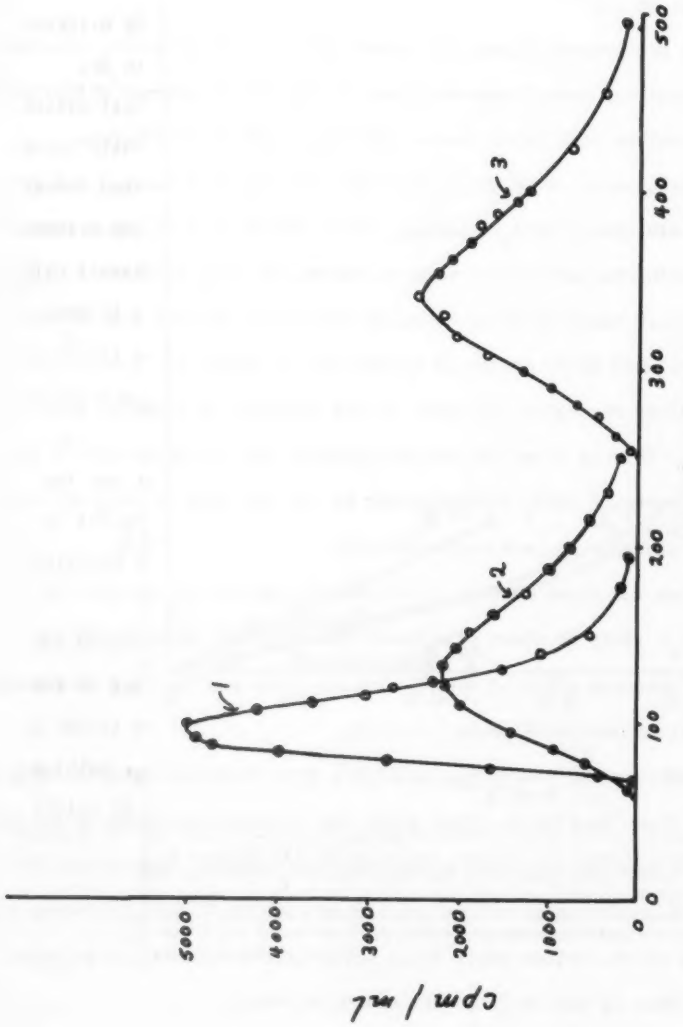

FIGURE 3. The spreading and displacement of narrow bands of trace concentrations of Sr-89 during the leaching of Al-soils or Al-montmorillonite with 10^{-3}M $CaCl_2$. Column capacities, in meq, were 0.8 for Davidson (1), 4.4 for White Store (2) and 8.0 for montmorillonite (3). Note the variable "dispersion" indicated by peak widths.

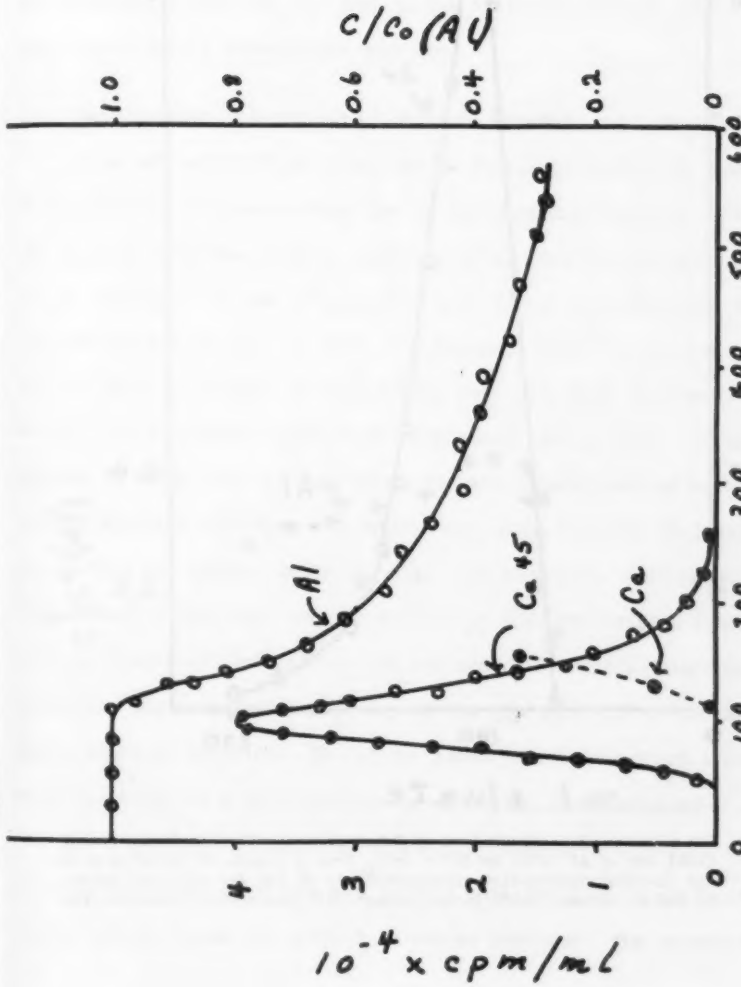

FIGURE 4. The elution of a narrow band of trace-concentration $CaCl_2$ (solid line) and of Al (dotted line) by 10^{-3}M $CaCl_2$ from a column of White Store soil, showing that the Ca tracer rides the front between Ca and displaced Al.

Note the variable "dispersion" indicated by peak widths.

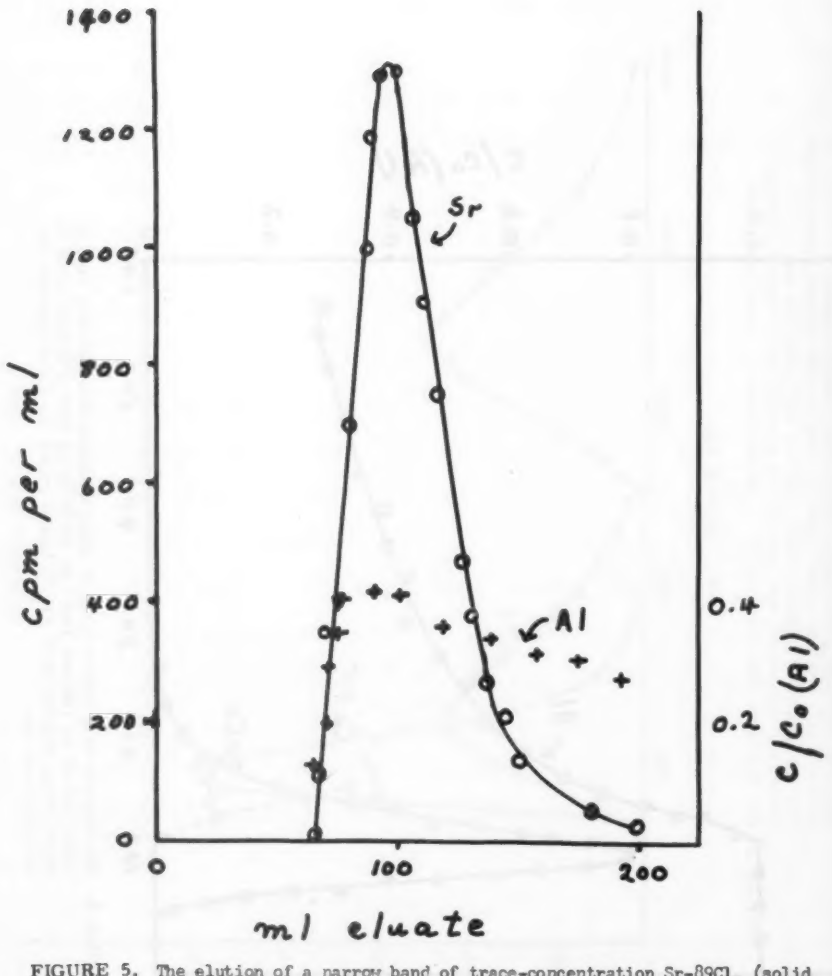

FIGURE 5. The elution of a narrow band of trace-concentration Sr-89Cl_2 (solid line) and of Al (x's) by 10^{-3} M $CaCl_2$ from a column of Davidson soil. The electrolyte-sorptive characteristics of the red soil are shown by the Al curve. Sr-89 broke through with percolating electrolyte.

When Sr-89 tracer is followed by $CaCl_2$ solution into soils saturated with various proportions of Al and Ca, the quantity of solution necessary to elute the Sr is greater the larger the proportion of capacity countered by Ca. This is shown for Cecil soil in Fig. 6. This behavior points up the obvious but often overlooked fact, that part of the capacity countered by an ion as strongly bound as Al^3 just is not available for the uptake of other ions from a dilute percolating solution.

III Thermodynamic Description of Exchange Sorption and Donnan Invasion

It is not difficult to write down a formal thermodynamic description of the sorption phenomena described in the preceding section, if we take into account only the minimal condition of electrical neutrality in sorbent and in solution. We see at once that this formal approach makes no interesting predictions for us. In fact, the approach would be quite sterile if it did not serve as a means of summarizing data and, more important, of indicating theoretical approaches which might make useful predictions. It is quite useless to write down the conditions for the general sorbent capable of primary sorption of anions and cations and which can also be invaded by whole electrolyte and solvent water as well. The equations would be nearly as complicated as the facts and there probably does not exist a single experiment to which they could be applied. We can summarize existing information and illustrate the necessary inadequacy of the approach with a discussion of simple exchange sorption. To fix our ideas we consider first a special case which has received fairly complete treatment in the laboratory[16].

The structural information on the clay mineral attapulgite suggests strongly that this substance may fulfill quite well the specifications of a simple ion-exchanger not subject to Donnan invasion. The accepted structure,

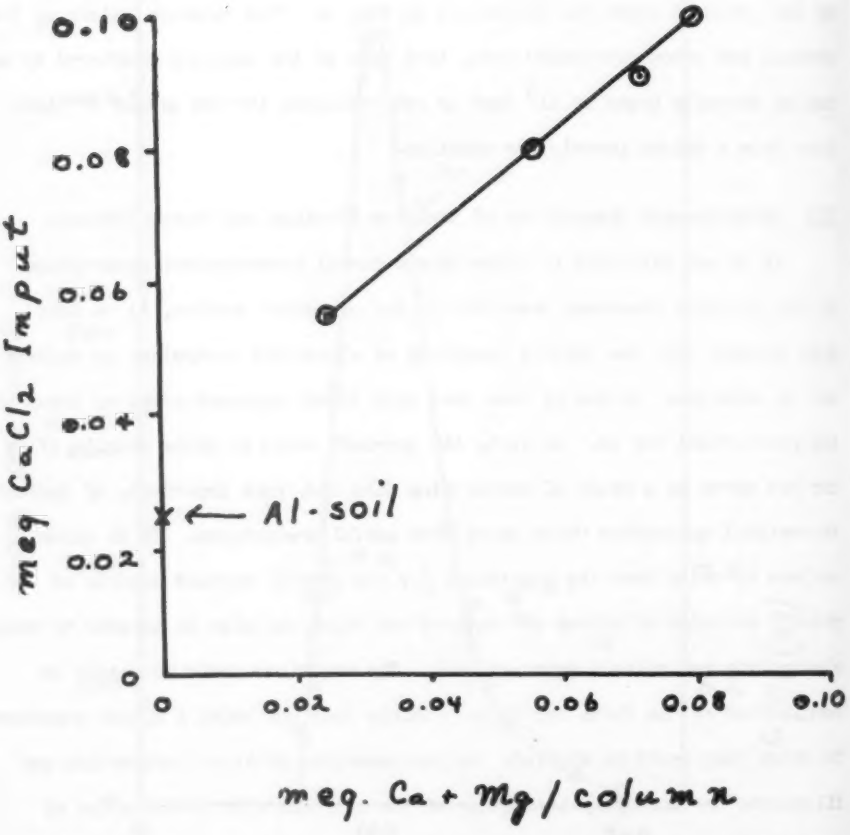

FIGURE 6. The quantity of displacing electrolyte (10^{-3}M $CaCl_2$) required to elute a narrow band of trace-concentration $Sr^{89}Cl_2$ from columns of red Cecil soil depends on the quantity of exchangeable Ca+Mg and is related reciprocally to the amount of exchangeable Al, which counters that part of the capacity not balanced by the alkaline earth ions.

proposed by Bradley[15], represents the material as consisting of an aggregate of narrow passages through a silicate matrix, in an arrangement similar to that of a honeycomb. The exchangeable ions are associated with the active sites deep within the tubules of the comb, and so, perhaps, are well removed from the direct influence of the surrounding solution. We now imagine a sample of this material with all the sites occupied by sodium immersed in a solution containing both cesium and sodium nitrates and the whole held at fixed temperature until all action ceases. Analysis will then show that a part of the sodium on the surface has been replaced by cesium. The condition that equilibrium is reached can be written in the form:

$$\frac{(Cs^+)\, f_{CsA}}{(Na^+)\, f_{NaA}} = K\, \frac{[Cs^+]\, \gamma_{CsNO_3}}{[Na^+]\, \gamma_{NaNO_3}} \qquad (1)$$

In this expression the symbols in parentheses indicate the fraction of the sites occupied by the designated ion; the bracketed symbols represent concentrations in solution. K is a true constant, depending only on the temperature. The γ's are the usual activity coefficients for the solution phase and the f's those for the solid phase. There is a certain arbitrariness allowable in the definition of the activity coefficients. Even when we accept the usual conventions for the γ's, those generally employed for solutions of electrolytes, we still have a certain freedom of choice in defining the f's. The choice will be dictated largely by the end in view. Thus we may choose a single standard state for each of the ions separately, in which case the value of K becomes identically unity and we obtain one set of f's which represent the data. Or we may wish, as we do now, to emphasize the dependence of f on the composition of the sorbent and choose pure sodium attapulgite and pure cesium attapulgite as reference states. For this choice

the details of the procedure for computing K and the f's have been given by Gaines and Thomas[17].

In Fig. 7 are given results of measurements on the Cs-Na-attapulgite system[16]. Here we plot values of lnK computed with complete neglect of the activity coefficients. It is evident that the simplified formula is entirely inadequate; we must allow a ten-fold variation in K to fit the facts. There is, of course, nothing surprising in this. The interactions of the ions with the clay surface must be quite strong and, furthermore, the ions on the surface are quite close together and undoubtedly interact with each other; we certainly expect to find non-ideal behavior on the surface.

A representation, in terms of the activity coefficients, of this non-ideal behavior for several clay-solution systems is given in Figs. 8, 9, 10. Our understanding of these systems reduces to our ability to produce a theoretical calculation of the behavior of the excess free energies, that is, the values of RTlnf. Little or no progress appears to have been made in this direction. While it is certainly true that the data are of insufficient accuracy to warrant any detailed discussion, the appearance of the curves for the activity coefficients suggests that we attempt to represent them according to the Margules expansions:

$$RT \ln f_1 = a_1(1-y)^2 + a_2(1-y)^3 + \cdots$$
$$RT \ln f_2 = (a_1 + \tfrac{3}{2} a_2)y^2 - a_2 y^3 + \cdots \qquad (2)$$

in which we write y for (Cs) in the cases given. We hasten to remark that this approach is entirely empirical, and quite devoid, so far as we are aware, of any theoretical justification. Thus for the exchanges between univalent ions we have (taking $a_2 = o$), for example:

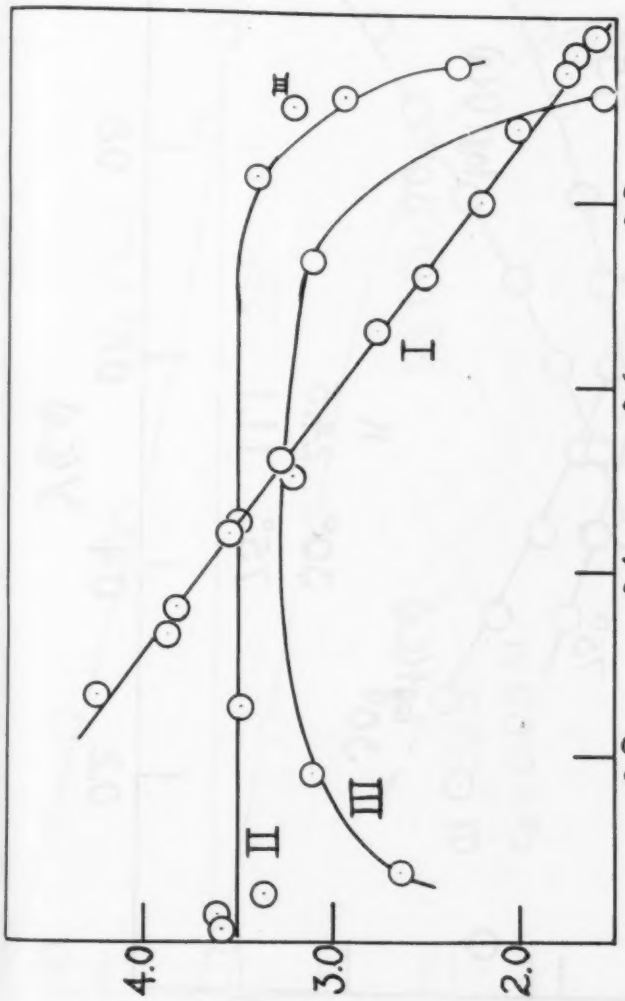

FIGURE 7. The varied behavior of the stoichiometric equilibrium quotient for the competition between cesium and ions of different valence types for sites on clay minerals. The logarithm of K_c is plotted as a function of the fraction of sites occupied by cesium, y(Cs). I, ln K for Cs-Na-Attapulgite; II, ln K-3 for Cs-Sr-Montmorillonite, 0.05N; III, ln K-4 for Cs-Y-Montmorillonite. Room temperature.

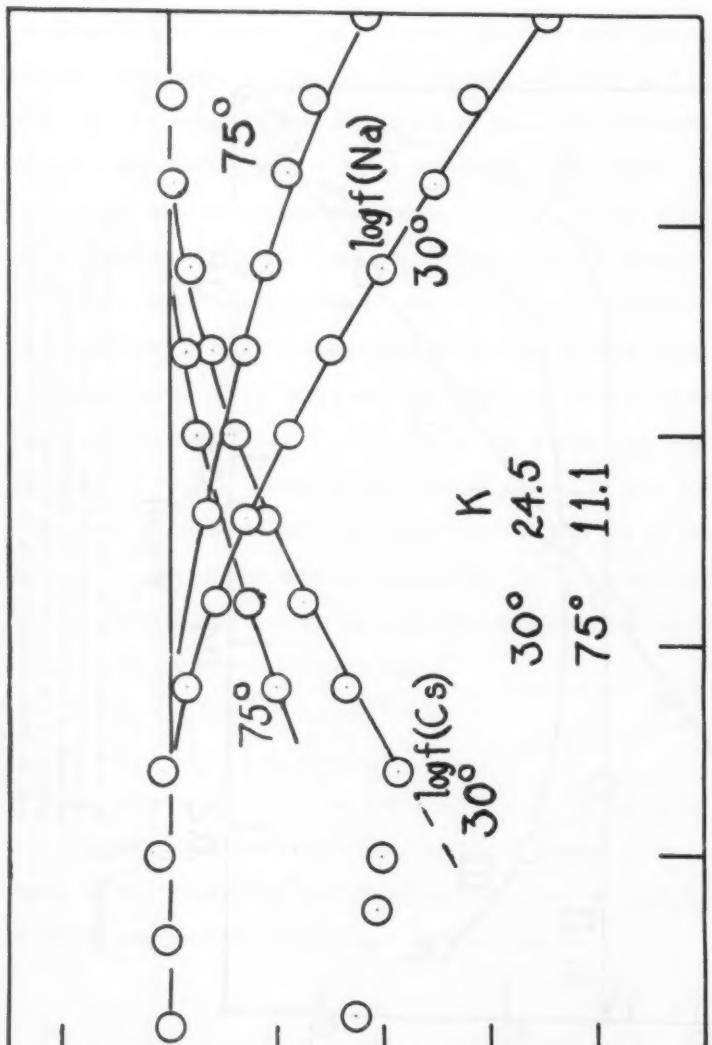

FIGURE 8. Thermodynamic functions for the system Cs-Na-Attapulgite.

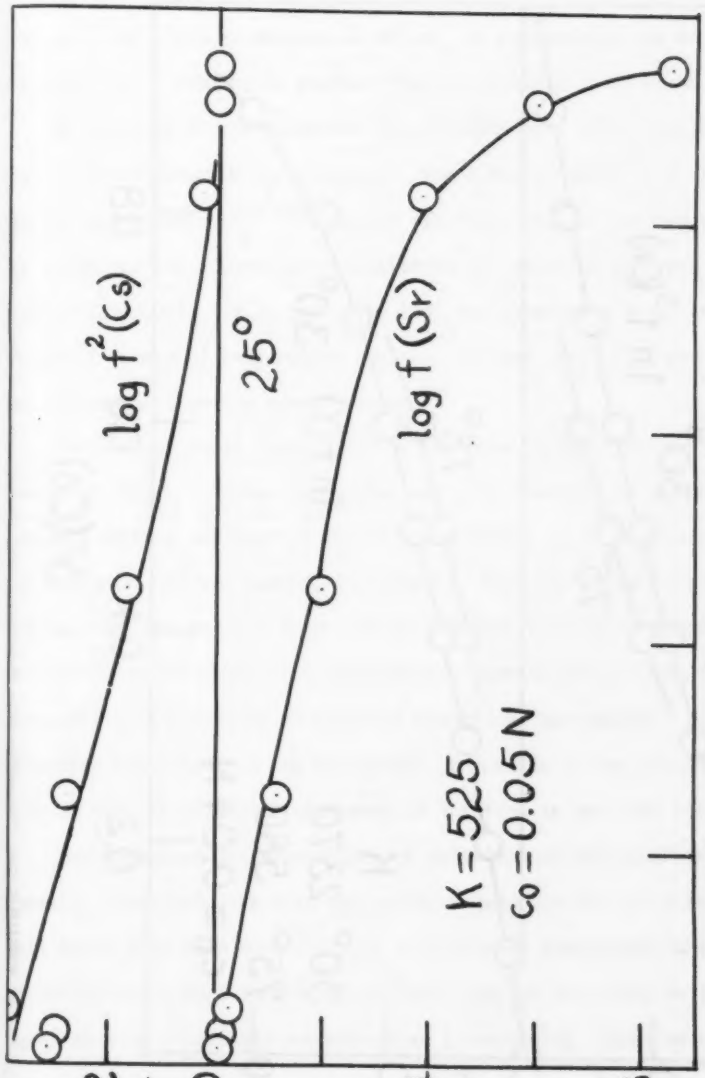

FIGURE 9. Thermodynamic functions for the system Cs-Sr-Montmorillonite.

FIGURE 8. Thermodynamic functions for the system Cs-Na-Attapulgite.

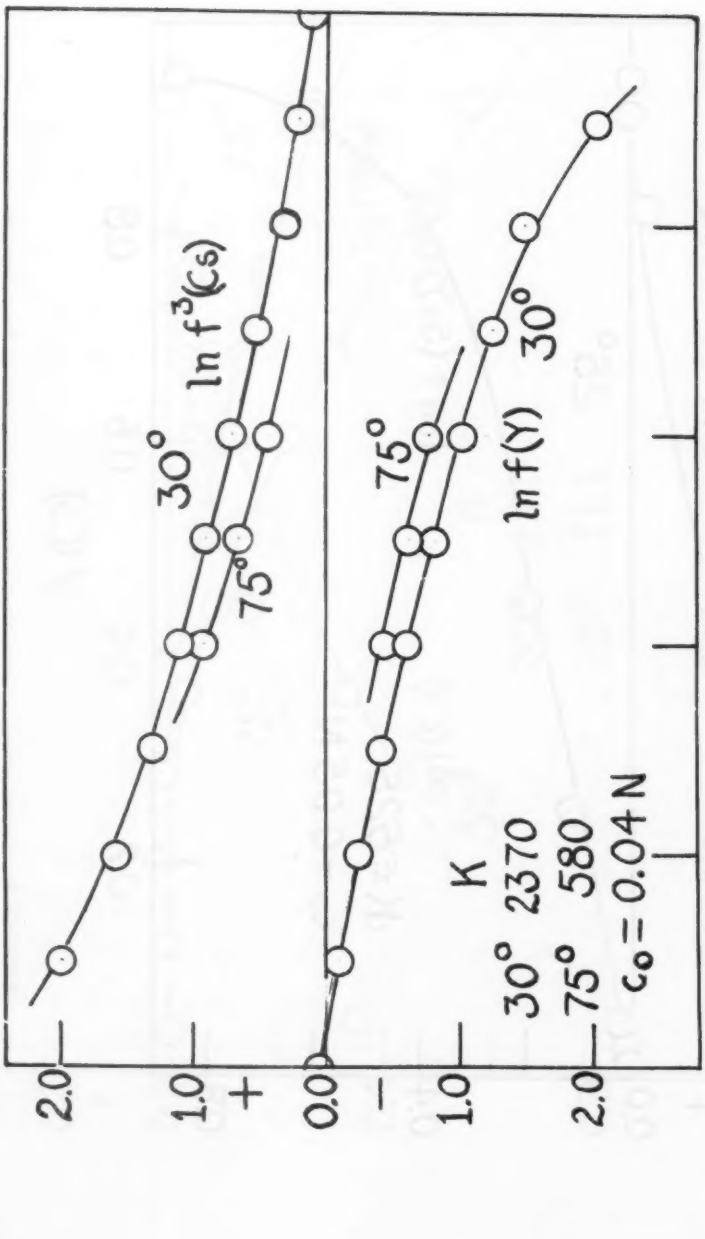

FIGURE 10. Thermodynamic functions for the system Cs-Y-Montmorillonite.

$$\frac{(Cs)e^{\frac{a_1}{RT}(1-y)^2}[Na]}{(Na)e^{\frac{a_1}{RT}y^2}[Cs]} = K \quad \text{or} \quad K_c = \frac{(Cs)[Na]}{(Na)[Cs]} = K e^{-\frac{a_1}{RT}(1-2y)} \qquad (3)$$

This suggests a linear dependence of $\ln K_c$ on composition, as is indeed found for this case. Nothing is further from the truth for the mixed valence types.

It would be highly desirable to establish some valid theoretical treatment of these activity coefficients. The limiting value of K_1 for y = o (in the above case: $K_c' = e^{-a_1/RT}$) gives the dependence of the cesium content of the sorber on the cesium concentration in the solution at trace levels: $\lim K_c = (Cs)/[Cs]$. It is just this limiting dependence which determines the motion of traces of radioactive nuclides through beds of simple soils under the influence of moving ground water.

The thermodynamic description of the case in which the sorbent is devoid of charge sites and consists only of a "sponge" of hydrous oxide involves nothing different from the distribution of an electrolyte between two solvents. We may expect, for example, that the interactions of the cations with the oxide matrix will be stronger than their interactions with water. There will then be a positive sorption of electrolyte, anions accompanying the cations to maintain electrical neutrality. It would not be generally expected that the proportion of cations in the sponge would be the same as that in solution; preferential sorption is the rule not the exception.

The thermodynamic description of this type of sorption can be made formally identical with that for exchange sorption discussed above. As is well known from much work with the resinous ion exchangers, the sorption of water may be of more importance in this case; in any complete treatment the water must be explicitly recognized as a component. Since we now do not

FIGURE 10. Thermodynamic functions for the system Ca-Y-Montmorillonite.

have a definite number of exchange sites, the activities of the sorbed species are best measured as molalities, m, referred to the amount of sorbed water. In the treatment of this case a single set of reference states is usually chosen so that the equilibrium conditions are of the form (for ions of charges ν_+ and ν_-)

$$m_{m(s)}^{\nu_+} m_{x(s)}^{\nu_-} \gamma_{\pm}^{\nu} = m_{m(e)}^{+} m_{x(e)}^{-} \gamma_{\pm}^{\nu}(e) \qquad (4)$$

The subscripts (s) and (l) referring to the sorbed and liquid phases, respectively. It might at once be remarked that this formalism would serve equally well for the mixed cases, invasion plus exchange on fixed sites. Generally, however, with soils and soil minerals the direct determination of the water content is either extremely difficult or quite impossible and the thermodynamic formalism based on exchange sites is more useful. The difficulty is not avoided but is, rather, segregated in the equations. It has been possible in cases where little or no electrolyte invasion was present to show that variations in water content are unimportant[17].

IV Motion of Sorbed Materials in Soils

The heterogeneous character of the crust of the earth, both on large and small scale, needs no further comment. Any remarks we make here on the motion of sorbed fission fragments in the soil must be taken as presupposing the best possible mapping of the relevant subterranean motion of water. We shall make no attempt to enlarge upon the difficulties of this undertaking except to remark that such a mapping adequate for our purposes is probably possible only through the use of the most advanced tracer techniques, utilizing helium and tritium as tracers for soil water. We shall assume in the following brief summary a great deal of knowledge: namely, that we

know the direction and average rate of flow of soil water in any locality of interest to us. In fact, we really must assume more than this. When a great bulk of solution is introduced into the soil, the local flow characteristics of ground water are changed and we must suppose that we have some means of learning what changes take place. Worse still, the introduction of chemical solutions in the earth will generally change the porosity and other hydrodynamically important properties of the soil. In reality, then, we are presupposing the continual application of tracer techniques to keep us completely up-to-date in our knowledge of the flow of ground water. Such a project is not at all beyond the realm of possibility on a limited scale. W. J. Kaufman and his co-workers[18] have successfully demonstrated the feasibility of several reliable tracers for ground water in actual field tests. Assuming that we have this detailed information, then, we can proceed to illustrate how it may be used in conjunction with the equilibrium data for sorption.

In the problem of tracing the fate of long-lived fission wastes in the earth, we shall generally be dealing with rates of flow so low that effects of diffusion may well become of importance. To illustrate the essential features of the problem we write out the differential equation which expresses the conservation of matter when a zone of material of concentration c, sorbed to the extent q is moving downstream both by convection and diffusion:

$$D\frac{\partial^2 c}{\partial x^2} - R\frac{\partial c}{\partial x} - \lambda\frac{\partial c}{\partial t} = \frac{\partial q}{\partial t} \qquad (5)$$

The applicable coefficient of diffusion, supposed constant, is D; something of the character of the soil is expressed by the porosity λ; and R is the linear rate of convective flow of the fluid. Much remains to be done in specifying what is meant by the "applicable" coefficient of diffusion. This quantity

depends in a complicated manner on the characteristics of the porous medium through which the liquid is flowing. Generally, values of D which are required to describe approximately the dispersion of solute during its progress through porous materials are much larger than molecular diffusion coefficients.

It is to Scheidegger[19/,20/,21/] that we owe the most ambitious attempts at a theoretical treatment of the dispersion effect as it applies to non-interacting solute. Working equations, obtained from a solution of the differential equation (5) with appropriate boundary conditions, are:

For a slug of non-interacting solute being displaced in one dimension by flowing solvent

$$c/c_o = \frac{X_o}{\sqrt{4\pi Dt}} \exp\left\{-(x-Rt)^2/4Dt\right\} \qquad (6)$$

xo is the initial thickness of the slug, x is the distance down the column under consideration, t is time and R is the linear velocity of the liquid. Scheidegger calls D the "factor of dispersion". Apparently it is a number which describes more or less approximately the fact that all of the liquid flowing through a porous medium does not move with the same velocity or traverse the same distance as it threads its way downward through tortuous channels interspersing the solid framework.

For the feed of solution of uniform concentration into a column whose pores are occupied initially by water, the shape of the boundary is given by:

$$c/c_o = 1/2 - 1/2 \operatorname{erf} \frac{x - Rt}{\sqrt{4Dt}} \qquad (7)$$

where the symbols have the same meaning as before.

Equation 6 says that the concentration profile for an initially narrow slug of solute will take the shape of a normal error curve as the material

is swept down a column. The area under the curve is always the same because the amount of solute remains the same. The height diminishes and the curve broadens as the slug is swept downward. The larger D, the factor of dispersion, the greater will be the broadening of the slug. Similarly, equation 7 tells us that the leading edge of percolating solution will attain a concentration profile which is sigmoid in shape and which becomes more and more slanting as time goes on. At a particular depth, the diffuseness of the front depends on D, being greater for larger values. Several recent studies have been made of the dispersion effect. Much significant work has come from the group in the Sanitary Engineering Department at Berkeley. In the doctoral dissertation of M. N. E. Rifai[22] are presented detailed studies of the effect of flow rate on the dispersion coefficient D for different types of porous beds. In this work, however, clean sands with nearly zero exchange capacity were used, corresponding to $dq/dt = o$ in equation 5. Other pertinent experiments performed by the Berkeley group have been reported by Orcutt, et al.[14] and by Kaufman and Orlub[18].

We are concerned mainly with two features of the dispersion process as it applies to non-interacting solutes. One has to do with the abilities of equations similar to 6 and 7 to describe column operation. Apparently this requires only the choice of an appropriate value for D. The second is more difficult, since it deals with inquiries into what such an appropriate value should be. Patently, it is impossible to treat this matter satisfactorily at present. All we can do is point out some of the characteristics of the factor of dispersion as gleaned from the experimental results of others.

D varies with the nature of the porous material through which flow occurs and with the flow rate. It always is found to be much larger than molecular diffusion coefficients, as evidenced by the fact that dispersion invariably

is greater than can be accounted for by ordinary diffusion. For example, Day[23] found values of D ranging from 10^{-5} to 8×10^{-3} for sand, soil and cation exchange resin columns.

Another characteristic of D is that it appears to change linearly with rate of flow. Orcutt, et al.[14] found the shape of the breakthrough curve (Cl through sand) to be independent of flow rate when the latter varied between 5×10^{-4} and 1 cm sec^{-1}. Day[24] found that a quantity , called by him the "index of dispersion" and given by: $f = 2D/V$, was a column characteristic, independent of flow rate, which could be inserted into equations 6 or 7 to describe boundary shapes during leaching.

Gardner and Brooks[25] ran water through homogeneously saline soil and sand columns and analyzed effluent concentrations. Boundaries between salty and salt-free water in eluates were diffuse, more so the longer the columns. Spreading of the boundaries was accounted for as a hydrodynamic dispersion effect in a way analogous to the treatment of non-equilibrium ion exchange chromatography. Salt was regarded as being in two categories, "mobile" and "immobile". The former was regarded as being in main channels through which water flowed downward, the latter in lateral channels or clogged pores. Following Thomas[26] and Vermeulen and Hiester[27] they wrote down conservation equations and indicated graphical methods for their solution, after the insertion of two column characteristics, the ratio of "immobile" to "mobile" salt (B) and the mobilization rate constant, (a).

The equation describing desalinization is:

$$c/co = (1 - J(s,t)), \qquad (8)$$

where s and t are dimensionless parameters:

$$s = afz$$

$$t = a(V-fz)/B$$

fz is the pore volume and V is the quantity of water which has entered the surface of the soil. Graphs of J(s,t) vs t/s are given. The column constants, a and B, are empirical, being chosen to give the best fit to data. For a collection of 18 soil or sand columns, B was between 0.064 and 0.364, while a varied from 0.19 to 3.27 cm^{-1}.

Very little work has been done in cases for relatively rapid flow in which sorption is an important factor. In any such study another serious complication will arise: For high flow rates we cannot suppose that the sorption processes go to equilibrium and we are forced to introduce a rate function for dq/dt. The resulting problem becomes most formidable, and it appears doubtful if detailed studies of this type will prove to be of much practical importance. Such studies, however, are necessary as aids in the interpretation of laboratory work designed to produce information on the equilibria involving the fission products.

The general character of the dispersion which occurs in ion-exchange systems can be illustrated by Fig. 11, which shows the breakthrough of Sr-89-labeled $SrCl_2$ for two columns of Sr-saturated soil. For isotopic exchange without dispersion or other non-equilibrium effects, vertical rather than slanting curves should be obtained. In actuality, Sr-89 began to leak long before a volume of solution corresponding to column capacity had passed through. Further examples of the dispersion of sorbed ions may be found in Fig. 3, 4 and 5, presented earlier.

In the study of the motion of sorbed material in the soil, two essentially equivalent approaches are open to us. We start by supposing that the basic information on the equilibria is available. Since we will generally be dealing with trace quantities in the chemical sense, this information will consist of a collection of sorption isotherms in the trace region. Presently

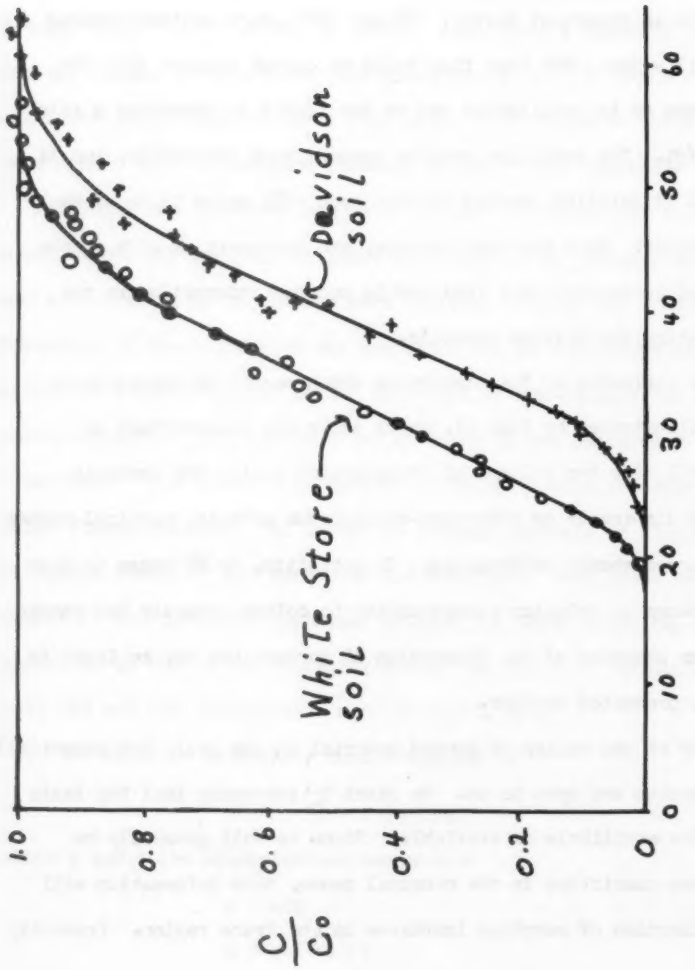

FIGURE 11. The breakthrough of Sr-89-labeled 10^{-3} M $SrCl_2$ for columns of Sr-soil (White Store on the left, Davidson on the right). The slanting breakthrough curves indicate the kind of dispersion effects which occur in the course of ion exchange in flowing systems.

available information on this subject is meagre. In particular, it is not clear that we may generally expect simple proportionalities between concentrations of sorbed species and the concentrations in solution. If these isotherms are significantly curved in the trace regions, a variety of effects may appear, and it will be extremely difficult to differentiate between these and the effects of dispersion and of slow rate of diffusion in the sorbents. The first approach would be to "adjust" the differential equation above to fit the facts as nearly as they are understood and, probably relying on machine computation, use the resulting equation in making the required predictions. As a highly over-simplified example (which would not require a computer for its solution), suppose that the rate of flow of soil water is so low as to be negligible, and that q = kc is the required sorption isotherm, then

$$\frac{(D\alpha)}{\alpha+k}\frac{\partial^2 c}{\partial x^2} = \frac{\partial c}{\partial t} \qquad (10)$$

and we have only a simple diffusion problem to contend with. In this case, however, we require a knowledge of the diffusion coefficient in the soil in question. The details of making this measurement have been worked out[28]. It is by no means obvious that the required value of D is that coefficient pertaining to the solution in question and that the only modification is the decrease brought about by the blocking effect, through α, and the partial immobilization, through k. This problem needs further attention; part of the diffusion might well be from site to site in the soil, an effect not explicitly cared for in the above equation.

Another, and more important, limiting case is represented by the situation when R is sufficiently large so that the term in D is negligible and we may still assume equilibrium sorption. We then have

breakthrough curves indicate the kind of dispersion effects which occur in the course of ion exchange in flowing systems.

$$R\frac{\partial c}{\partial t} = -(\varkappa + k)\frac{\partial c}{\partial t} \qquad (11)$$

the well-known equation for equilibrium chromatography. Application of this equation will give perhaps all the required information in many cases. In particular, however, it cannot give any details as to the shapes of zones of varying concentration. Thus it is necessarily incapable of accounting for "leaks" of low concentrations of, say, radio-strontium (Fig. 11). A knowledge of the possibility of such leaks is highly desirable.

The second possible approach is to disregard entirely all the mechanistic details and treat the problem according to methods similar to those developed for distilling columns. This whole approach, utilizing the idea of the equilibrium plate, has recently received considerable development at the hands of A. S. Said[29]. The method is necessarily entirely empirical in character. Its application to the prediction of the motion of radioactive nuclides in the soil would depend on the completion of fairly detailed preliminary studies carried out under similar conditions. This approach may prove to be the most practical. A survey of a proposed ground disposal site might well consist of a mapping via tracers of the flow of ground water. In the process numerous representative samples of soil would be obtained. Equilibrium sorption measurements on these under conditions envisaged for the disposal operation would give much of the required information. In any case, since all of these somewhat academic approaches can never be more than guides in practical situations, it will undoubtedly be best to explore the applicabil of all or several of them in conjunction with the necessary monitoring operations.

V Summary of Some Pressing Problems

It is strikingly apparent from the survey presented in this paper how thoroughly entangled several apparently unrelated scientific disciplines have become in the problem of tracing the fate of radionuclides in the earth. The most complex problems of hydrodynamics are made worse by some of the most subtle difficulties in surface chemistry, and both are in turn complicated by the necessity of applying the results to conglomerates of our most impure chemical compounds: soils. Fortunately it is not necessary to wait for the solution of all of these difficulties before we can proceed to practical problems. We always have the laboratory as the best recourse for quick answers to specific questions. It is surely true, however, that less time will be wasted in the laboratory, as well as in the field, the better the theoretical guides we have available. We would like to conclude our survey, then, with a mention of some of the directions in which we feel that scientific work might most profitably proceed.

Very much of importance remains to be done in identifying and characterizing the different kinds of surface activity exhibited by the soil minerals. The classification given in this paper is far too general to do more than serve as a guide in specific problems. Thus, the investigation of the complex mixtures of hydrous oxides and clay minerals has scarcely been started; much remains that should be done in this field, particularly as regards the sorption of anions and the effects thereby produced.

Although much has been done in the general theory of sorption, we badly need more fundamental theory as a guide. More realistic models for exchange sorption are required to give reasonable interpretations to the very varied types of results that are found with the soil minerals. It is to be hoped

that a really competent theoretical chemist will interest himself in these problems.

We need, as we have already suggested, a more thorough analysis of the flow problems relating to the motion of fluids in sorbing systems. Although it is clear that many of the mathematical problems here involved are far too complex for explicit solution, with the availability of modern computing machines practical answers to the questions can still be obtained. It is in the formulation of these questions that there is much need for more definitive laboratory work together with careful and intelligent analysis of the results.

Of the many specific problems which need attention we conclude by mentioning only one. Especially in attempts to keep track of cesium and strontium in the earth, we need a thorough investigation of the problem of "fixation" on soil minerals, the conditions under which it takes place and its differentiation from cases of exceptionally strong adsorption. The solution to this problem alone might well save much useless effort in tracking cesium in the earth and perhaps may suggest other and practical means for waste disposal.

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A STUDY OF THE MIGRATION OF RADIOELEMENTS IN SOILS

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The growing use of atomic energy for peaceful purposes has resulted in the production of a greater volume of nuclear fuel, and this inevitably leads to an accumulation of radioactive waste the bulk of which is intended for interment. The problem of how to process and store the waste of radiochemical production becomes of increasingly greater importance to the national economy and may eventually limit the further scope of the use of atomic energy.

Several methods of interring radioactive waste have been described (1-12):

1. The storage of steamed solutions in underground steel or concrete reservoirs;

2. Encasement in glass or cement blocks with the subsequent burying of blocks in the earth or dumping them into the ocean;

3. Drainage of radioactive solutions into the ocean or rivers;

4. Dumping of radioactive waste into closed basins and other earth depositories.

5. The use of pits and old oil wells.

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The cheapest and most accessible method should be the method of interring radioactive waste in open earth depositories. However, the problems of the sorption of fission products by minerals in soils and in the earth have not yet been adequately studied (13-21). Besides, in spite of the use of protective screens and colmatage layers the seepage of radioactive solutions into the underlying soils is always possible. The conditions under which radioisotopes will migrate with subsoil waters have been very inadequately analyzed until now (22-24).

The present paper is devoted to a study of the laws governing the sorption of uranium fission products by some natural sorbents, such as soils and soil forming minerals, from solutions with a varying salt content. Particular attention is paid to strontium-90 as the most dangerous isotope from the physiological standpoint. Experiments were made both in the laboratory and in the field. Among the materials used were alkaline carbonate and acidic non-carbonate sands containing humus, carbonate clay loam, light carbonate clays, medium carbonate clays, a number of clayey minerals, feldspars, the mineral groups of calcites, gypsum, etc.

Two types of solutions were studied:

1) An alkaline solution containing NaOH (4-8 gm/1) and $NaNO_3$ (200 gm/1);

2) An acidic solution, containing HNO_3 (6-8 gm/1) and $Al(NO_3)_3$ (200 gm/1).

Radioelements, carrier-free or with carrier, were added to the initial solutions, or a mixture of uranium fission products was introduced.

Static or dynamic methods were used in the studies. In the static experiments, the quantities of sorbent were shaken with definite portions of the solution before equilibrium was reached. After this, radiometric analyses were made. The dynamic experiments were carried out in glass cylinders with a diameter of 1cm filled with various quantities of sorbent. Previous to this, the sorbents were ground and a fraction of 80-100 mesh was selected although some of them, like montmorillonite, kaolinite, etc., had a much higher dispersion. In order to study the sorbing properties of soils, quantities of from 3 to 400 gms per litre were used.

Laboratory tests proved that the sorption of all the known radioelements, with the exception of radiozirconium, radioniobium and radiocaesium, is much more complete from alkaline solutions than from acidic.

The $_{p}H$ of the medium had a particularly strong effect on the sorption of cerium. Thus, up to 98–100 per cent of cerium was absorbed from an alkaline solution containing sodium nitrate, regardless of the initial concentration of cerium (interval of concentration-from imponderable quantities up to $3.3.10^{-4} \div 3.3.10^{-3}$ mg-eq/l, or the quantity or nature of the soil (quantities of soil from 3 to 30 gms per litre of solution). As for acidic media, from solutions containing aluminium, cerium was practically not caught up at all by the soils.

The degree of sorption of radioruthenium was extremely low, but in an alkaline solution it may reach several tenths of millicurie per 100 gms of soil when cationic ruthenium is used. Practically no ruthenium was absorbed from an acidic medium. Anionic ruthenium was absolutely not absorbed by soils.

The sorption of strontium in an acidic medium was several times lower than in an alkaline medium. When an alkaline solution with a strontium concentration of $5.32.10^{-3}$ mg-eq/l was used, the soils, regardless of their nature, taken in quantities of 30 gms per litre absorbed roughly 60 per cent of the strontium. Under similar conditions only 5–6 per cent of the input quantity was absorbed from an acidic aluminium solution. With a reduction of the quantity or soils tenfold, the sorption of strontium in an alkaline medium falls to 30–33 per cent.

Apart from this, a great number of natural sorbents, mainly pure minerals of inorganic origin, making up the earths and soils (25) was tested to determine to what degree they absorb microquantities of strontium. The following solutions were used in the experiments in this stage of the work (Table 1).

Table 1

No.of Solution	Activity for $Sr^{89,90}$ curie/litre	Concentration of Calcium Ions, mg/litre
1	$1.5.10^{-6}$	0
2	$1.5.10^{-5}$	0
3	8.10^{-6}	100

The first two solutions contained only imponderable quantities of $Sr^{89.90}$ (10^{-7} - 10^{-9}H) of varying levels of activity without the addition of any salts whatsoever. Calcium chloride was added to the third solution, so that the concentration of calcium was approximately equal to its content in river water (26).

The results of the experiments are given in Tables 2 and 3. The montmorillonite clays of various origin are denoted as follows: A - montmorillonite clay from Kzyl-CP (Kazakhstan), B - Oglanlin (Turkman Republic); C - Crimea (Kil); D - Askangel (Georgia); The $_{p}H$ value of solutions, equal in weight, fluctuated from 7.2 to 9. The quantity of sorbent equalled 1 gram per 50 millilitres of solution.

The Sorption of Radiostrontium by Some Natural Silicates and Alumosilicates

Table 2

Group	Mineral	No.of Input Solution	Percentage of Radiostrontium Absorbed
1	2	3	4
Hydromica	Glauconite	1	83
		2	78
		3	70
	Vermiculite	1	88
		2	91
		3	82
Mica	Biotite	1	72
		2	81
		3	72
	Muscovite	1	87
		2	85
		3	80

Table 2 (ctd)

5	6	7	8
Montmorillonite Group	A	1	99.9
		2	99
		3	98
	B	1	99.9
		2	99.9
		3	99
	C	1	92
		2	95
		3	76
	D	1	98
		2	99
		3	92

9	10	11	12
Quartz Group	Quartz	1	20
		2	19
		3	6
Nepheline syenites (rock)	Mariupolite	1	86
		2	92
		3	54
	Hibinite	1	70
		2	72
		3	29
Nepheline Group	Nepheline	1	71
		2	69
		3	40

1	2	3	4
Serpentine-Kaolinite Group	Serpentine	1	66
		2	63
		3	36
	Kaolinite	1	68
		2	67
		3	40
	Dickite	1	38
		2	29
		3	20
Talcum Pyrophillite Group	Talcum	1	18
		2	21
		3	15
	Pyrophillite	1	14
		2	14
		3	6
Disthen Group	Disthen	1	44
		2	44
		3	22

Table 2 (ctd)

5	6	7	8
Ceolites	Natrolite	1	60
		2	66
		3	36
	Desmine	1	22
		2	27
		3	12
Halloysite Group	Halloysite	1	88
		2	89
		3	73
Pyroxene Group	Egyrine	1	50
		2	60
		3	21
	Diopside	1	47
		2	47
		3	19
Amphiboles	Hornblende	1	45
		2	48
		3	23

9	10	11	12
Asbestos Group	Asbestos	1	56
		2	48
		3	41
Feldspars	Albite	1	27
		2	32
		3	5.5
	Oligoclase	1	39
		2	37
		3	24
	Labradorite	1	50
		2	61
		3	35
	Microline	1	28
		2	24
		3	11

Table 3

The Sorption of Radiostrontium by Various Minerals Other than Silicates and Alumosilicates

Group	Mineral	No. of Input Solution	Percentage of Radiostrontium Sorption
1	2	3	4
Calcite Group	Magnesite	1	34
		2	33
		3	24
	Calcite	1	40
		2	48
		3	21
	Limestone (rock)	1	45
		2	49
		3	39
	Aragonite	1	39
		2	40
		3	27
	Iceland Spar	1	33
		2	38
		3	20
5	6	7	8
Materials of an Organic Origin	Peat	1	93
		2	92
		3	75
	Peat Slag	1	59
		2	54
		3	39
	Brown Coal	1	60
		2	58
		3	37
	Diatomite	1	58
		2	60
		3	39
Covellite Group	Covellite	1	2
		2	4
		3	1

Table 3 (ctd)

1	2	3	4
Calcite Group	Ankerite	1	35
		2	40
		3	15
	Dolomite	1	30
		2	28
		3	21
Gypsum Group	Anhydride	1	44
		2	39
		3	25
	Gypsum-Alabaster	1	48
		2	50
		3	16
Rutile Group	Pyrolusite	1	99.9
		2	99
		3	95
Corundum-Ilmenite Group	Hematite	1	28
		2	20
		3	22

5	6	7	8
Lepidocretagoethite Group	Limonite	1	50
		2	48
		3	35
	Hydrogoethite	1	70
		2	63
		3	49
Apatite Group	Phosphorite	1	96
		2	94
		3	89
	Fluor-apatite	1	61
		2	59
		3	41
Hydrargillite Group	Hydrargillite	1	58
		2	57
		3	39

The above tables (2 and 3) show that among the substances tested, those that absorb radiostrontium best are montmorillonite, kaolinite, micas, hydromicas, pyrolusite, halloysite, peat, phosphorite and nepheline syenites. These sorbents also have the greatest sorption capacity (27).

It should be noted that the mineral groups very common in earths and soils, such as calcite, quartz, gypsum, feldspar,etc, absorb radiostrontium to an insignificant degree.

Thus, the sorption of radiostrontium by the inorganic part of soils and earths is mainly determined by clayey minerals which usually make up the colloidal dispersion fraction of the abovementioned substances. Besides, as Tables 2 and 3 make clear, the presence of calcium ions in a concentration approximately equal to their content in river water, considerably reduce the sorption of radiostrontium by the sorbents analysed. It may be expected that radiostrontium once it gets into the earths and soils, will, everything else being equal, to a greater degree be held up where clayey minerals are present and where subsoil waters contain the least quantity of calcium.

Our experiments failedto reveal that the level of radioactivity has any effect on the degree of sorption of radiostrontium. This is apparently due to the fact that comparatively large quantities of sorbents have been used.

The sorption of radiostrontium by montmorillonite was the subject of more comprehensive studies. The mineral in question is very common both in a dispersed state as well as in the form of large deposits (27). A study was made of Strontium-90 sorption by montmorillonite clays (deposits A, B, C, see p.4) from solutions containing varying concentration of ions: H^+, Na^+, K^+, NH_4^+, Ca^{+2}, Mg^{+2}, Ba^{+2}, Fe^{+3}, Al^{+3}. The ionic strength of the solution during the experiments remained constant, with pH being equal to 7-8. In the study of the behaviour of microquantities of Strontium in the sorbent-solution system, depending on the composition of the solution, the method of determining the distribution coefficient (γ) in static conditions was used (28):

$$\eta = \frac{VC_{me} \cdot 100}{VC_{me} + q_{me}}, \qquad (1)$$

where V - volume of solution,

C_{me} - an equilibrial concentration of the ion under study in a solution,

q_{me} - ion quantity, absorbed by a given quantity of sorbent.

Fig. 1 shows the results of the measurement of radiostrontium distribution coefficient for montmorillonite from the Oglanlin deposits. The curves reflecting the change in the distribution coefficient depending on the composition of the solution for montmorillonite from the two other deposits are insignificantly displaced with respect to each other. However, the nature of the changes, as well as their sequence, in case of varying cations present in the solution together with the radiostrontium, remains constant for all three sorbents. This is quite understandable, considering that they are minerally indentical. Consequently, the nature of montmorillonite has an effect on some of the quantitative parameters characterizing sorption without altering the process of sorption as a whole. That is why, for the sake of economizing space, only data reflecting the process of the sorption of radiostrontium by the montmorillonite from the Oglanlin deposit are given. According to the position of the curves in Fig. 1 the cations for montmorillonite may be arranged in the following series, according to their ability to reduce the sorption of radiostrontium: $Al^{+3} > Fe^{+3}$, $Ba^{+2} \geq Ca^{+2} > Mg^{+2} \geq H^{+} > NH_4^{+} > K^{+} > Na^{+}$.

The above series makes clear that, in general, the higher the cation valency - and in case of equal valency, the smaller the radius of the hydrated ions - the greater its effect in reducing the degree of radiostrontium sorption by montmorillonite. It should be noted that the results pertaining to the $Sr^{+2} - Fe^{+3}$, $Sr^{+2} - Al^{+3}$ systems should not be compared with similar data for monovalent and bivalent cations because of using solutions with pH=2 in experiments.

Our calculations have shown that the process of radiostron-

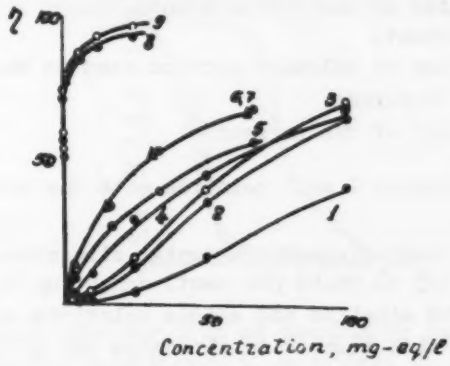

Fig.1. Curves showing the distribution of micro-quantities of strontium between solutions and sorbents in the presence of various cations: 1.Na^+; 2.K^+; 3.NH_4^+; 4.H^+; 5.Mg^{+2}; 6.Ca^{+2}; 7.Ba^{+2}; 8.Fe^{+3}; 9.Al^{+3}.

tium sorption by montmorillonite, being an ion exchange one, is quantitatively described with great precision by the law of mass action. It should be noted that there is no basic difference in the sorption of strontium in macroareas and microareas of the concentration of its ions. The exchange constants received with weight quantities of strontium may be suitably used to calculate the process of sorption of traces of strontium.

The desorption of radiostrontium from montmorillonite is also subject to the law of mass action which has been used to work out the formula allowing the calculation of the quantity of radiostrontium remaining in the sorbent after "n" treatments of it by portions of desorbing solution with a constant salt composition:

$$q_n = q_0 \left(\frac{k}{\sqrt{\frac{V}{m}} + k}\right)^n , \qquad (2)$$

where q_0 - the quantity of radiostrontium on the sorbent prior to desorption,

q_n - the quantity of radiostrontium on the sorbent after n desorption events.

n - number of desorption events,
k - constant,
v - volume of solution portion used in the desorption of strontium,
m - weight of the sorbent.

Calculations on Formula 2 well coincide with the results of the experiment.

Of the other radioelement fragments, zirconium and niobium are equally well (up to 98-99 per cent) caught up by the soils analyzed, both from alkaline and acidic solutions as well. The capacity of soils with respect to zirconium and niobium has not been established for it is, apparently, extremely great while it is impossible to add macroquantities of zirconium and nioboum into the solution due to the low solubility of the compounds of these elements in the solutions used.

The degree to which caesium is absorbed by soils depends very little on the acidity of the medium. From solutions with a caesium concentration of $1,2.10^{-3}$ mg-eq/l with the use of 30 gm/l of soil, sands absorb 50 per cent, sandy loams 90 per cent, clays 90 per cent of the input quantity.

The maximum capacity of soils with respect to caesium is established with the aid of dynamic tests in glass columns. The filtration speed is 0.5-1.0 ml per hour. It has been discovered that sandy loams absorb 4 mg-eq of caesium per 100 gms of soil, with 11 per cent of the quantity absorbed easily washed away with water. Sandy loams catch up 1.25 mg-eq of caesium per 100 gms. The subsequent treatment of the sandy loam with water will wash away up to 30 per cent of the caesium. However, the soil is capable of absorbing from a fresh caesium solution quantitites equal to those washed away.

Fig.2 shows the curves of the sorption (1) desorption (2) and secondary sorption (3) of caesium. In the experiment, an alkaline solution with a 10 mg-eq/l of $CsNO_3$ has been used. The quantity of sandy loam in the cylinder was equal to 10 grams. The results indicate that the curves of desorption and secondary sorption are practically identical.

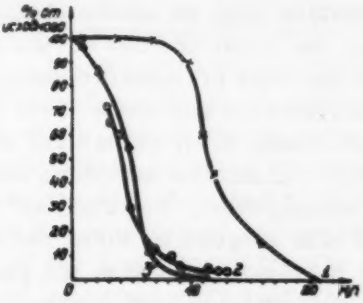

Fig.2. The sorption of caesium by sandy loam in dynamic conditions. 1. sorption; 2. desorption; 3. secondary sorption.

The testing of the sorption properties of soils with respect to a mixture of fission products in alkali and acidic solutions confirmed the preliminary results of experiments in which single radioelements have been used. The solutions used in these experiments have the following radiochemical compositions: Sr^{90} 14.5 per cent; Cs^{137} 16 per cent, Ru^{106} 68 per cent, rare earth elements 0.5 per cent, Zr^{95} + Nb^{95} 1 per cent with the total β-radioactivity of about 50 millicurie/litre.

The study of the sorption property of sands has been conducted in dynamic conditions with a filtration speed of 2-5 ml per hour. It has been established that in an alkali medium the filtration of radioactivity takes place in the first drops of the filtrate due to the presence of the low-sorbing ruthenium- not more than 30 per cent of the input quantity of ruthenium in the solution is retained by the sand.

The capacity of non-carbonate sandy loams containing humus has, with respect to the total number of elements used, 39-48 mg-eq. radium in γ-radiation and 410-538 millicurie in β-radiation per 100 grams of soil. The capacity of carbonate sand is somewhat lower, and stands at 10 mg-eq, radium in γ-radiation and 180 millicurie in β-radiation per 100 gms of soil.

There is practically no sorption of radioruthenium, rare

earths and radiostrontium when an acidic aluminium solution is passed through sand. The first portion of the filtered solution containes about 100 per cent of radioruthenium and rare earths, 80 per cent of radiostrontium and about 15-18 per cent of radiozirconium and radioniobium. Thus, in natural conditions, sands will absorb from acidic aluminium solutions only radiocaesium, radiozirconium and radioniobium. The study of the sorption properties of sands with respect to these radioelements in an acidic medium is of no interest because the preliminary experiments with the several radioelements have revealed that they are equally well absorbed from an alkaline as from an acidic medium.

The sorption property of sandy loams and clays with respect to alkaline solutions has been studied under static condition. A quantity of soil has been treated with solution until there is manifest sorption of radioactivity from the solution. It has been discovered that radiostrontium and radiocaesium are absorbed from a salt alkaline solution in the following quantities:

No.	Soil	Radiocaesium Millicurie/ 100 gms	Radiostrontium Millicurie/ 100 gms
1.	Medium Sandy Loam, Carbonate	242.0	7.0
2.	Light Clay, Carbonate	200.0	9.4
3.	Sand, Carbonate	172.0	9.4

The sorption of radiozirconium, radioniobium and rare earths has not been determined since it may be expected, on the basis of preliminary experiments, that in an alkaline medium the capacity of soil with respect to them is very great. The content of these elements in the solutions analyzed is insignificant.

The sorption of radioelements will apparently largely depend on the thickness of the soil layer through which the liquid filters. Laboratory experiments with alkaline solutions contai-

ning fission products have shown that with the increase in the quantities of soil - the solution volume being constant - the degree of the sorption of a mixture of radioelements goes up to a certain limit which depends on the presence of the non-sorbing anionic radioruthenium. By using quantities of soils up to 400 gm/l for the purification of a radioactive alkaline solution, the degree of the sorption of radioelements may be raised to 94-96 per cent of the input quantity. The further increase in the quantities of soils does mit increase the degree of sorption (Fig.3).

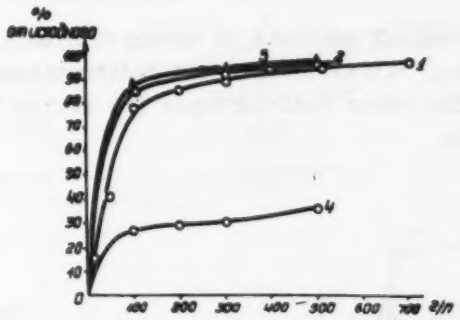

Fig.3. Relation between the sorption of the sum of element fragments and the quantity of soil. Sorption from alkaline media. 1. Heavy carbonate sandy loam; 2. light carbonate clay; 3. medium carbonate sandy loam.
Sorption from acidic medium: 4. medium carbonate sandy loam.

From an acidic solution, regardless of the quantity of soils, the degree of sorption of the sum of the radioelements is extremely small and reaches 28-30 per cent. In general, radiozirconium, radioniobium and, to a certain extent, radiocaesium are absorbed from a strongly acidic medium. When soils which have absorbed radioisotopes are treated with water or an

alkaline solution, a part of the radioelements pass into the solution. The washing with a salt solution is much more effective than with water:

Soil	Conditions of Desorption	Radioisotopes Washed Off (in per cent of sorbed activity)
Carbonate Sandy Loam	Alkaline Solution	3.0
	Water	0.2
Light Carbonate Clay	Alkaline Solution	4.0
	Water	0.14
Sand, Carbonate	Alkaline Solution	4.13
	Water	0.14

The radiochemical analysis of desorption solutions reveals that, in the main, radiostrontium and radioruthemuim are washed off the soils. The other radioisotopes are rather firmly retained by the soils.

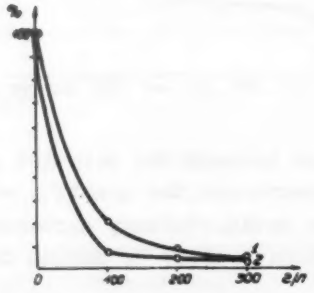

Fig.4. Secondary sorption of the sum of radioelements from desorbing solutions. 1. sorption from an alkaline solution; 2. sorption from water.

The desorbed elements are again caught up by the soils with the degree of secondary sorption depending on the quantity of added soil. Fig.4 shows the dependance of the reduction in the radioactivity of the washing off solution up on the quantity

of soil.

Thus, the laboratory experiments indicate that in the soils the most mobile are strontium and ruthenium.

In order to check the capability of soils in natural surroundings, to absorb and retain the fission products from solutions and subsoil waters, field experiments were staged. For this purpose a plot was used whose main surface soils were alluvial sands, mostly fine-grained, with insignificant layers and lensings of clay loams. In the thick of the sands, at a depth between five and seven metres, there were subsoil waters whose movement had a laminary character. From above and below the water-bearing layer was limited by a dense layer of sandy loam of small thickness. Preliminary studies had been made to determine the hydrogeological characteristics of the plot and the direction and speed of the movement of the soil stream.

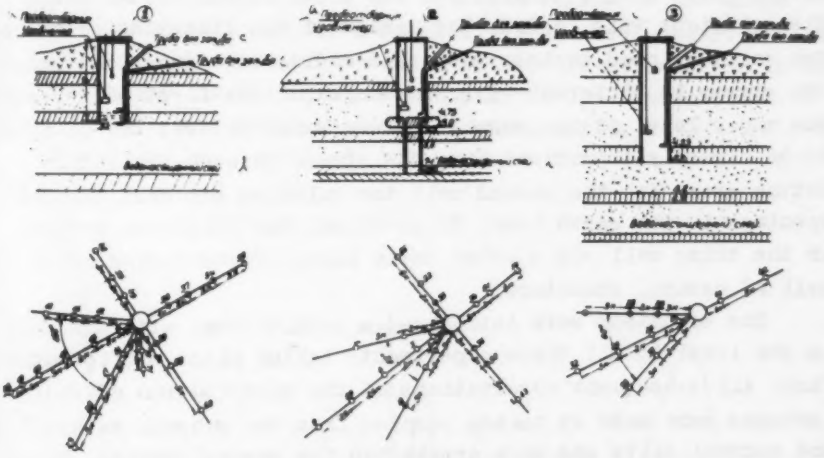

Fig.5. Construction of wells and arrangement of control slits. 1. Well No. 1; 2. Well No. 2; 3. Well No. 3
a) Sampler; b) Level gauge; c) Input pipe;
d) Water-saturated layer.

Three experimental filtering wells had been sunk on the plot (Fig.5) through which the solution containing fission products and sodium nitrate was introduced into the soil stream. Three hundred curie of radioactive substances with a sodium nitrate of 200 gm/l was thrown into each well. The radioactive solutions contained Sr^{90}, Cs^{137}, Zr^{95}, Nb^{95}, Ru^{106}, Ce^{144} and other fragmentary rare earth elements.

The construction design of the wells and the position of the control slits shown in Fig.4 was designed to study the distribution of radioactive isotopes in the waterbearing layer in the direction of the soil stream as well as against it. The wells were chinks one metre in diameter inlaid with asbocement piping and covered with bitumen on the inside. Each well had a cover, level gauge, sampler and two drain pipes through which the solutions were loaded. The design of the filtering bottom of the wells allowed to introduce the radioactive solutions into the stream in different ways. The depth of the first well reached the upper level of the subsoil waters which allowed the solution to be directly introduced into the stream through the entire bottom area. In the second well the solution was shed through openings in the earth lock, 25 cm thick. The filtering bottom in the third well was a metre thick layer of underlying sandy soil of natural structure.

The solutions were introduced a single time, with the fall in the level in all three experiments taking place sufficiently fast. All subsequent observations of the distribution of radioisotopes were made by taking samples from the subsoil waters from the control slits and were carried on for several months. Piezometers on the slits were mounted in such a way as to have the perforated part of the piping cover the waterbearing soil layer. Before each sample was taken, the water in the well was carefully stirred and was kept for settling for three to five minutes.

The pH value was determined in all the samples of water directly. This was determined to fluctuate between 7+8 for the entire period of the experiments. The radiometric and radiochemical analysis demanded preliminary concentration, and that was effected mainly by steaming down to no less than 10 times.

The observations have showen that the distribution of radioactive pollutions in the waterbearing layer may be divided into two periods. The first period pertains to the time when the solutions penetrated into the subsoil waters through the wells, when the natural movement of the soil stream is disrupted. In that period, due to the slow movement speed of the subsoil waters and the fast filtration of the solutions their flow in the stream take place practically over a similar radius in every direction. An increase in beta and gamma radioactivity of the water both along the stream as well as against it has been determined in the slits nearest to the wells. It had been continueing for several days after the solution was poured in and was recorded within a radius of three metres from the first and second wells (Figs.6 and 7), and within a radius of one metre from the third well. At the same time, it had been observed that the level of pollution of the water in the direction opposite to the stream was, in the first six days after the introduction of the solution, somewhat higher in all three wells.

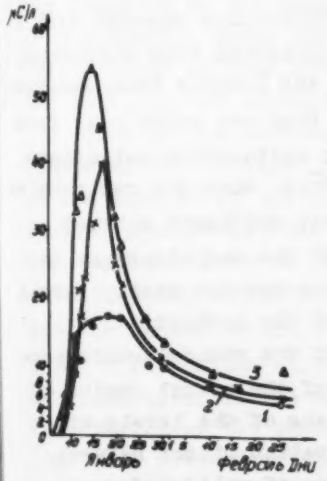

Fig.6. The content of the total β-radioactivity in the slits of well No.1. 1. Slit No.12; 2. Slit No.13; 3. Slit No.14.

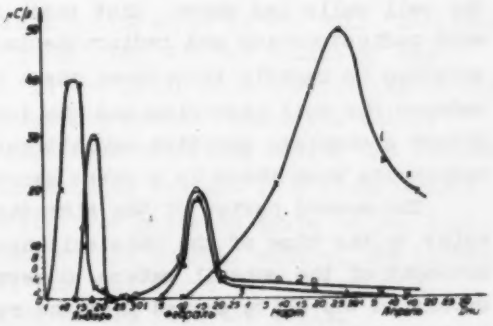

Fig.7. The content of the total γ-radioactivity in the slits of well No.2. 1. Slit No.3; 2. Slit No.4; 3. Slit No.5.

However, the increase in the degree of radioactivity of the water in the slits situated along the stream continued considerably longer than in the slits arranged in the opposite direction. Thus, the radioactivity of the water in slit 1 increased in the course of 12 days, in slits 11-20 days and in slit 24-35 days; meanwhile, the fall in radioactivity in the slits of the opposite direction (37,38 and 41) began on the sixth day of the operation of the wells.

Thansk to the effect of pressure and the high filtrating qualities of the well bottoms, the distribution of the radioactive elements in the stream took place over the entire depth of the water bearing layer. The time of contact between the soil particles with the solution proved to be insufficiently long for the easily sorbed radioelements (zirconium niobium, caesium and the rare earth elements) to be fixed in the soil. In that period of time, the water in the slits nearest to the well (at a distance of one metre from the well wall) contained all the fission products introduced into the well with a slightly altered percentage content of radioelements in comparison with the input solution.

According to laboratory tests, the filtration through such a quantity of soil should have ensured purification from zirconium, niobium, cerium, the rare earth elements and largely from caesium. Analyses of the waters in the slits more than one metre away from the well walls had shown that their main radioactive pollutions were radiostrontium and radioruthenium. Thus, when the radioactive solution is rapidly introduced there is not sufficent contact between the soil particles and the ions of the radioisotopes to effect a complete sorption equilibrium even for the easily sorbed components when there is a great excess of the sorbent.

The second period of the migration of the radioelements pertains to the time of the reestablishment of the normal regime of movement of the subsoil waters. Observations of the levels of waters in the slits showed that the restoration of the natural regime of the stream took place in the area of wells and approximately after three days and after some 6-8 days in the area of well. It was approximately then that the level of radioactivity of the water in the shafts arranged against the stream ceased to

rise. The migration of the radioelements in that period depends only on the processes of sorption and desorption on the water bearing soils and the stationary speed of movement of the subsoil waters.

As a result of the observations, we established the systematic rise and fall in the radioactivity of the water in the slits in the direction of the movement of the subsoil stream. The slits of the first radius (one metre from the well wall) revealed a single maximum for the level of radioactivity, while for the slits of the second radius (three metres from the well wall) there was a twofold rise in the radioactivity of the water (Fig.7). The first maximums were mainly due to the outflow of the solutions in the initial period, whereas the second were the result of the migration of radioelements along the stream under the normal movement of subsoil waters. As the area of the water polluted by radioactive substances advanced there was a marked fall in the level of radioactivity.

In the first radius of the slits the pollution of the water was measured by the tens and hundreds of millicurie per litre; in the second, three metres of the well, the radioactivity had fallen three or four orders, and in the third, five metres from the well, it was another order lower.

Test drills were made on the polluted area to study the nature of the sorption of the radioisotopes by the soil. Analyses of the samples of the soils taken from various points of the aquifer showed that the fall in the level of radioactivity on the area between slits of the first and second radius was mainly caused by the sorption of radioelements on the soils. The further decline in the level of radioactivity was due to a considerable dissolution in the stream which took place in spite of the laminary character of the movement of the subsoil waters. The latter fact served as the cause for limiting our observations to a radius of between seven and 10 metres from the wells.

With time, the radiochemical analysis of the pollutions migrating along the aquifer changed. Under the normal movement of the stream, zirconium, niobium, caesium and the rare earth

elements were sorbed and well retained by the water bearing sand, which served to substantiate our laboratory experiments. Radiocaesium was somewhat more mobile, but, in the main, the radioactivity of the water was determined by radiostrontium and radioruthenium (Figs.8 and 9). The advance of radiostrontium along the stream took place at a speed practically equal to the speed of the movement of the subsoil waters. Trace quantities of radiostrontium were found in the samples taken from slits seven metres away from the wells. Analyses of the soils taken at a distance of over three metres from the well revealed the absence of radioactive pollutions in them.

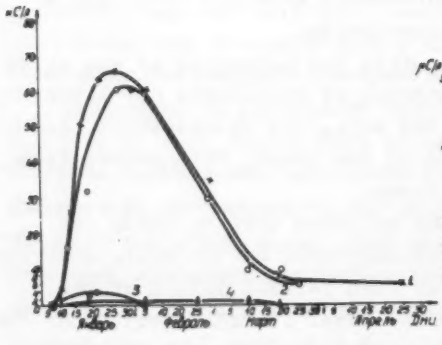

Fig.8. The radiochemical analysis of the polluted water of slit No.11. 1. Total ß-radioactivity. 2. Ru content; 3. Sr Content; 4. Cs content.

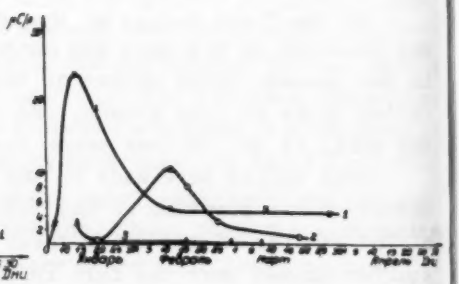

Fig.9. Radiochemical content of water contamination, Slit 13 (Sr-content 1, Ru - 2, Cs - 3).

Thus, the data obtained testifies to the very difficult sorption of radiostrontium in water bearing soils and the easy move-

ment of the element in subsoil waters.

Our observations of the migration of radioruthenium have allowed us to establish the fact that the speed of its distribution in the stream is roughly twice as slow as that of radiostrontium.

The ease with which radiostrontium is desorbed from sandy soils of natural structure have been additionally checked in the process of washing the underlying bottom of the third well. Naturally, the presence of a bottom monolith in that well as a filter resulted in the sorption of a considerable part of the radioelements introduced, and reduced the radius of their distribution in the first period of the operation of the well. To check the strength of the retention of radioactivity on the soil, a washing of the monolith was made at the end of the experiment with a 4 m^3 acidulated solution of sodium nitrate (HNO_3 2 gm/l, $NaNO_3$ 200 gm/l). In the process of washing it was established that it was mainly radiostrontium that was washed off the soil and distributed along the stream. Of the other elements only radioruthenium was observed. Radiocaesium, washed off the sandy bottom of the well was retained by the neighbouring layers of the soil.

Thus, the results of field experiments proved the basic data of the laboratory research on the laws governing the sorption of fission products by soils. The fact that the experiments were carried out in conditions least favouring sorption and the fixation of the radioactive substances (the stream of subsoil waters) went to prove that the interment of such radioelements as zirconium, niobium, cerium and the rare earth elements should not give rise to serious apprehension. It appears that the behaviour of caesium is also favourable but requires additional study.

The main danger in the drainage of radioactive waste in earth depositories is unquestionably presented by strontium, due to the considerable desorption of it even by subsoil waters. Anionic ruthenium is likewise practically not sorbed by soils of various mineralogical composition. From that point of view, the

areas most favourable for the construction of earth depositores are those which have low-lying subsoil waters with slow movement, low content of calcium ions and generally low salination.

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TREATMENT OF HIGH-LEVEL WASTES BY SINTERING

By

T. Tamura*, Oak Ridge National Laboratory

One method under investigation for treating high-level wastes is the fixation of fission products in highly insoluble fused ceramic masses. Amphlett and co-workers use soil as the base of the ceramic material.[1] The Canadian investigators utilize nepheline syenite to supply the soda, alumina, and silica.[2] Professor Patrick of Johns Hopkins University produces crystalline silicates to retain the fission products.[3]

The purpose of fixation treatment is to produce a waste form which will minimize environmental contamination by having the fission products embedded in a durable and insoluble solid. It is not difficult to prepare fused, durable ceramic sinters or glasses by employing temperatures of approximately 1300° C.[4] It would be desirable to prepare these durable

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1. Amphlett, A. B., and Warren, D. T., "Fixation of Activity in Solid Form by Absorption on Soils," Part II, AERE C/R 2202, 1957.

2. Durham, R. W., "Report of Working Meeting on Fixation of Radioactivity in Stable, Solid Media at the Johns Hopkins University," June 19-21, 1957, TID 7550, p. 8., 1958.

3. Patrick, W. A., Ibid., p. 3.

4. Dickson, J. H., "Glass," Hutchinson's Scientific and Technical Publications, London, 1951.

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sinters or glasses at lower temperatures of about 850° C. A lower temperature of firing would minimize the volatilization of some fission products such as cesium.

With lower heating temperatures in mind, studies were made of sinters which would retain a large percentage of the fission products. In order to limit the parameters, the waste used was simulated neutralized Purex prepared according to the method outlined by Lacy.[5] Since the fission element most difficult to fix is cesium, the studies were primarily concerned with the leachability of this element. The cesium concentration was kept at 100 mg per liter in the simulated waste solution; cesium 137 tracer was used for the analytical determination of cesium. Although several different techniques were used to determine the amount and rate of leaching, the most reliable technique was found to be the method recommended by the Committee on Durability of the American Ceramic Society.[6]

Early Experiments on the Production of Sinters. Although earlier experiments were subject to errors of judgment, they were valuable in establishing certain ideas and in gaining information on the operational techniques of sinter production. It is interesting to note that the early experiments showed the necessity of using over 60 g of illite per 50 ml of

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5. Lacy, W. J., "Manual for the Preparation of Simulated Fuel Reprocessing Waste Solutions," ORNL CF-58-4-45, 1958.

6. Morey, G. W., "The Properties of Glass," Reinhold Publishing Company, New York, 1954.

waste solution to produce a sinter which would retain cesium at a level of 99% or greater, even though 20 or 40 g of illite formed sinters strongly resembling glasses in contrast to more porous sinters.

In addition, information was sought through the literature. One of the more applicable reports was that by J. Enss.[7] Selected data from his report are summarized in Table 1. These data show the durability of glasses with the optimum amount of substituting cation (Table 1). The report also carries a range of concentrations for each cation. These data suggest that smaller and more highly charged cations are more effective in increasing the durability of sodium silicate glasses. Of particular interest is the effect of alumina on the durability of glasses, since clays are not only sources of silica but alumina as well.

Effect of the Form of Alumina on Cesium Retention by Sinters. Enss showed that alumina increases the durability of glasses; his study was conducted at temperatures much higher than 850° C and for durations of time which insured glass formation. With lower temperatures and limited heating time, it was felt that the form of alumina would influence the sinter's ability to retain cesium. In the use of clays as the additive, alumina is introduced into the system bonded with silicon through a common oxygen atom. If alumina were introduced without the common bond with silicon, it is likely that the soda would attack the silica and alumina independently, and a satisfactory sinter would not be formed.

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7. Enss, J., "Untersuchungen uber die Abhangigkeit der relativer Wasser-Angreisbarkeit Jes Glases von seiner Chemischer Zusammensetzung," Glasstech. Ber. 5, 449, 1928.

Table 1. Durability of Sodium Silicate Glasses Which Have Silica Substituted by Selected Oxides*

Components	Percentage of Components in Glass Number: 1	2	3	4	5
Na_2O	17.3	18.2	18.7	17.5	17.5
SiO_2	82.2	75.9	67.1	53.3	53.3
B_2O_3		5.3			
Al_2O_3			14.2		
CaO				28.7	
ZnO					28.4
Total Oxides ----------	99.5	99.4	100.0	99.5	99.2
Weight loss on leaching (%)	33.9	0.50	0.11	0.22	0.09

*Date from J. Enns.[7]

The results of several tests using clay and oxide ingredients are shown in Table 2. From columns 1 and 2, it is seen that the aluminosilicate (clay) is superior to the oxide ingredients. Comparing the results in columns 2, 3, and 4, one is led to conclude that alumina is not completely incorporated by the silicate; in fact, the formation of sodium aluminate is suggested by the high alumina content in the leaching solution. There is the further inference from these tests that the formation of durable sinters from $Al(NO_3)_3$ type waste would require a different treatment.

The Effect of Different Clay Minerals on Cesium Retention. The weight of clay used in these tests was based on the production of sinters of equal weight. Thus, clays with high moisture or hydroxyl content were added in greater amounts than clays with low moisture content. The clays which were selected represent the basic types of clay minerals found in soils and sediments. Furthermore, they represent structures which differ in the amount and manner in which aluminum is distributed. The bentonites represent structures with two separate layers of silicate; these silicate layers are joined together by an alumina layer. Kaolinite differs from the bentonite in having only one layer of silicate with the layer of alumina attached to it. The illite and Conasauga shale samples represent clays which have a similar arrangement as the bentonites; they differ in having aluminum in positions where silicon normally is found in the silicate layer. As much as one out of four silicon atoms may be replaced.

From the results shown in Table 3, and from earlier tests,[8] the

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8. Morgan, K. Z., "Health Physics Division Annual Progress Report," ORNL-2590, TID-4500, p. 77, 1958.

Table 2. The Retention of Cesium as Influenced by the Form And Amount of Alumina in the Mix

Composition	Grams Oxide in Sample Number: 1	2	3	4
Al_2O_3	20.7	20.7	2.50	2.07
SiO_2	41.0	41.0	54.9	41.0
Fe_2O_3	3.68	3.68	5.05	3.68
MgO	2.28	2.28	2.93	2.28
K_2O	4.88	4.88	6.61	4.88
Total Oxides	72.54	72.54	71.99	53.91
Sources of the Ingredients	Clay (illite)	Chemical (oxide or nitrate)	Chemical (oxide or nitrate)	Chemical (oxide or nitrate)
Cesium leached (%)	0.13	1.10	0.20	0.31
pH of leachate	----	----	11.20	11.80
Leachate composition (ppm):				
Na	46.0	28.1	80.0	250.0
Si	9.2	7.6	21.0	33.0
Al	3.3	165.0	20.0	60.0
K	2.0	55.0	34.0	73.0

Table 3. The Influence of Clay Type on the Retention of Cesium

Source and Material	Fithian Illite	Conasauga Shale	Wyoming Bentonite	Georgia Kaolin	Arizona Bentonite
Initial weight of clay used (g)	80.0	80.2	85.8	87.3	97.2
Final weight of sinter (g)	83.7	83.9	86.1	85.0	84.4
Cesium leached (%)	0.13	0.13	0.27	0.02	0.47
pH of leachate	10.4	10.0	11.1	9.63	10.7
Leachate composition (ppm):					
Na	50	34	180	15	65
Si	22	25	54	10	39
Al	5	6	7	4	5
K	8	3	1	1	< 1

bentonites are considered to be less effective than the other clays. The increased leachability of cesium from bentonites is thought to be due to the higher content of magnesium in these clays. Magnesium is present in place of aluminum in the structure of clay. Enss showed that aluminum is much more effective in increasing the durability of sodium silicates than magnesium; thus, when the clay is attacked by the sodium, the resultant silicate system containing magnesium would not be as durable as one containing aluminum. Available data are insufficient to draw final conclusions regarding the superiority of any one clay type.

Several interesting relationships became apparent and were verified. For example, it was noticed that if the sodium content in the leachate was high, the per cent or amount of cesium leached was high. This suggests a similar role for the two elements. Since the sodium content varies with different sinters, it is suggestive of an incomplete reaction, and the degree of completion affects the sinter's ability to retain cesium. In Fig. 1 the amount of cesium leached from four different sinter preparations is plotted as a function of the pH (a measure of the sodium content). The scatter diagram strongly suggests a significant relationship between the two parameters.

The Use of Boric Acid as a Fluxing Agent. If the inference regarding the incomplete reaction between the clay and soda is correct, then the use of a fluxing agent is indicated. Boric acid was selected since it is a stronger acid than silicic acid and because of its proven ability to improve durability according to Enss.[7] It was necessary to determine the amount of boric oxide which would accomplish the desired acceleration of fusion and

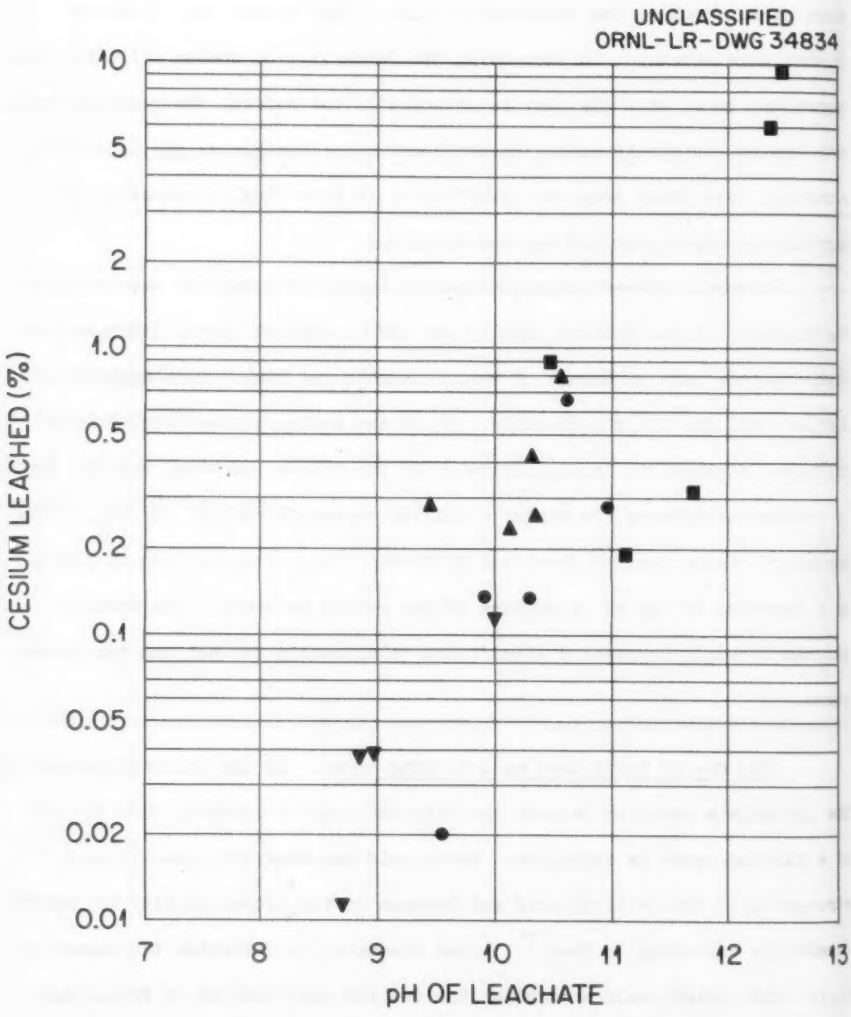

FIGURE 1. The relationship of the leachability of cesium and sodium from sinters. The pH is used as a measure of the sodium ion content.

yet not weaken the sinter by forming sodium borate. Table 4 shows the compositions of sinters prepared from Conasauga shale and different amounts of boric oxide. Note that the minimum of leached cesium is observed at 8.5% B_2O_3. At 17.1% B_2O_3 the sinter was well fused; however, chemical analysis showed that the silica and alumina were relatively more soluble. This indicates that the frame work has been weakened by an excessive amount of borate.

The addition of borates as an ingredient may be contrasted with the addition of alumina as an ingredient. It was shown earlier that high amounts of alumina resulted in a high concentration of aluminum leached from the sinter (Table 2). It might seem that a similar situation could result from the addition of borate. However, with as much as 17% B_2O_3 the leachability of cesium was below that found for the sinter containing no borate. The difference in the behavior of the two materials on the characteristic of the sinter lies in their "glass forming" or network forming parameters. The ability of elements to form glasses have been associated with the element's ionic size and charge. Elements, such as silicon, with a small size and high charge are particularly favored to form a three dimensional network which is the basic framework of glasses. Though both aluminum and boron have trivalent positive charges, aluminum is larger than silicon, whereas boron is smaller. This smaller size is favorable for the "glassy" network formation; and the boron appears to be easily incorporated within the silicate network. The latter statement is based on the observation that sinters containing as low as 2% B_2O_3 are massive and harder than those without borate indicating a well-integrated system for the borated sinters.

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Table 4. The Effect of Boric Oxide on the Retentivity Of Cesium by Sinters

	Sample No.			
	1	2	3	4
Weight % of:				
Conasauga Shale	87.9	83.6	79.3	70.6
B_2O_3	0.0	4.2	8.5	17.1
Na_2O	10.9	11.0	11.0	11.1
Cesium leached (%)	0.11	0.04	0.01	0.03
pH of leachate	10.0	9.0	8.8	8.9
Leachate composition (ppm):				
Na	38	22	1	4
Si	16	7	4	10
Al	5	4	2	8
K	3	2	1	4

Retention of Strontium by Sinters. Thus far no mention has been made of the retention of strontium by these sinters. The effort on cesium retention was prompted by earlier tests which showed strontium to be retained by sinters approximately ten times more effectively than was cesium.[9]

Since the ionic radius of strontium (1.13 Å) is similar to that of sodium (0.95 Å) and cesium (1.69 Å), they will all occupy similar positions in the aluminosilicate framework. Furthermore, it is common practice in glass technology to strengthen sodium silicate glasses by the addition of divalent ions such as zinc and calcium (see Table 1). Hence, evidence and reasoning point to a degree of retention for strontium which would be equal to or greater than for cesium.

A series of sinters with Conasauga shale and boric oxide (8.5%) were leached to determine the retention of strontium. The leaching of strontium, as measured by strontium 89 counting, was not detectable. The accuracy of the experimental method limited the establishment of retention at 99.99 ± 0.01%.

The approach to the fixation of cesium and strontium on a structural basis has been fruitful thus far. A similar approach to the sorption of these elements by clay minerals has also proved to be profitable. In contrast to the empirical techniques applied in the production of these sinters, sorption studies have been conducted on clay minerals whose structural parameters have been changed by selective treatment. These studies have shown that cesium sorption is particularly sensitive to changes in accessibility of the silicate framework.

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9. Morgan, K. Z., Ibid., p. 75.

Both sinter production and sorption studies with clay minerals are continuing. Earlier experiments are being repeated since the earlier leachi techniques were not reproducible due largely to difference in surface area of the sinters. Efforts to improve the sinter's ability to retain fission products and to produce sinters with higher bulk densities are in progress.

AN EXPERIMENTAL FACILITY FOR HIGH-LEVEL WASTE FIXATION STUDIES*

By

W. J. Boegly, Jr.**; and E. G. Struxness (p. 1844)

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One possible method for disposing of high-level radioactive wastes is the fixation of the fission products in a stable, solid medium. Several procedures and techniques have been and are being developed for "fixing" fission products in aluminosilicate solids with variations in the firing temperature and the method of firing. At Oak Ridge National Laboratory primary emphasis has been on the fixing of fission products and earthen materials at temperatures below 900° C using the fission product decay heat available in the waste solution. In recent months this development program has been broadened to include the use of supplementary heat from artificial sources in fixing wastes which do not contain enough decay energy to self-heat to 900° C.

Field experiments designed to simulate the heating expected in reactor fuel reprocessing wastes have shown that it is possible to obtain

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* Detailed Statement for the Congressional Record on Joint Congressional Committee on Atomic Energy Public Hearings on Industrial Radioactive Waste Disposal, January 28, 29, 30, and February 2, 3, 1959, Washington, D. C.

** Date of birth: April 25, 1931, Place of birth: Camden, New Jersey, Rensselaer Polytechnic Institute (1948-1952), B.C.E., Civil Engineering; Purdue University (1952-1953), M.S.C.E., Civil Engineering; Purdue University (1953-1956), Ph.D., Civil Engineering;
Barker & Wheeler (1952), Engineer; Purdue University (1952-1956) Research Fellow; Oak Ridge Nat'l Lab. Union Carbide Nuclear Co. (1956-present) Engineering Leader

some indication of the heat required to produce a satisfactory ceramic product, but data necessary for the evaluation of thermal gradients in self-heating and the evolution of radioactive off-gases cannot be obtained except by experimentation with actual radioactive waste solutions. These experiments can only be performed in a "hot" cell or in a suitable facility in the field. The design and construction of such a field facility for use in high-level waste-fixation experiments is the subject of this paper.

DESCRIPTION OF FACILITY

The experimental facility is composed of three parts: (1) the furnace and mixer; (2) the off-gas cleaning system; and (3) the devices for the thermal measurements and the power controls.

Furnace and Mixing Assembly

To insure the containment of fission products, the entire mixing and furnace assembly is located underground in a shielded concrete containment vessel (Figure 1). This provides for the isolation of all fission products, except those released to the off-gas system. The containment vessel is composed of two concrete liners: an outer liner, 10 feet in diameter and 10 feet deep, and an inner one, 6 feet in diameter and 9 1/2 feet deep. The annular space between the liners is filled with sand for shielding and structural purposes. It also provides a second barrier in case the inner liner fails. The mixer is located above the furnace and its contents are discharged to the furnace when a remotely-operated valve is opened. The furnace is composed of two stainless steel cylinders, an outer cylinder for structural

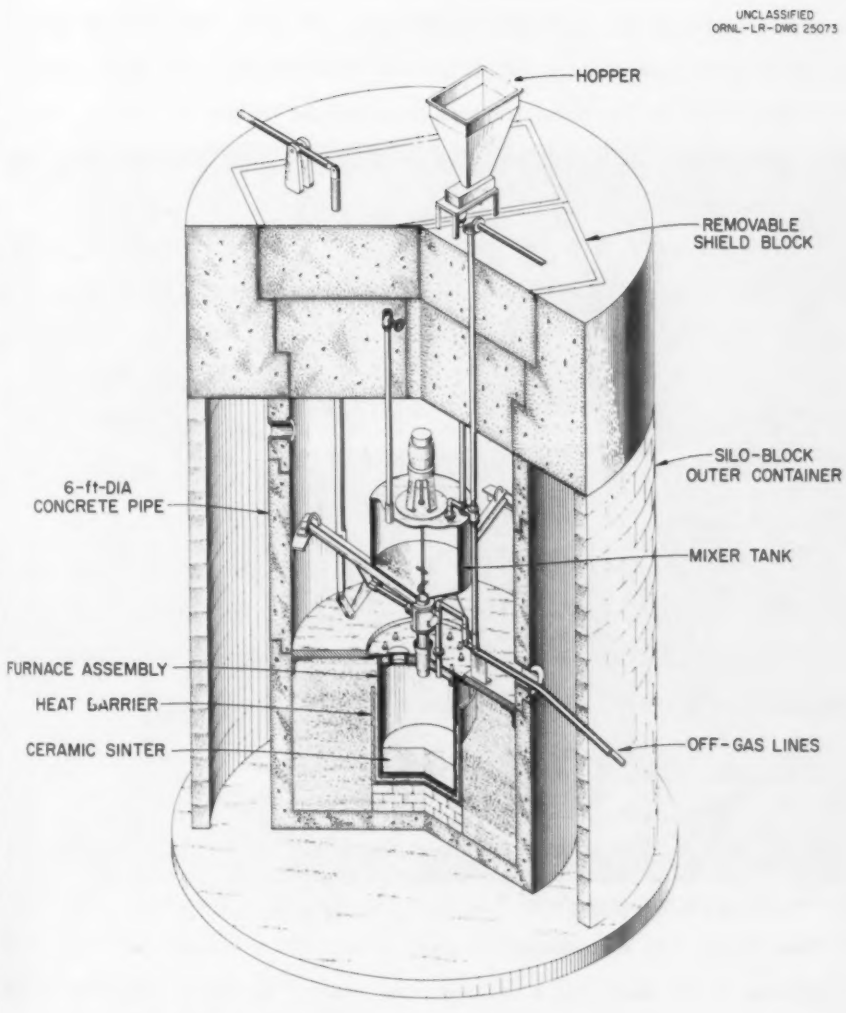

Fig. 1. Experimental Facility for High-Level Self-Sintering.

stability and safety and an inner cylinder to retain the fired sinter (Figure 2). An insulated cover, on which lifting hooks are provided for removal of the sinter, is bolted to the inner cylinder. The cover is equipped with an off-gas line, a slip-joint for the mixer discharge pipe, and connectors for the thermocouples used in measuring thermal gradients. All components which come in contact with the waste solution are fabricated from stainless steel.

The heat barrier, or "adiabatic heater," shown in Figure 3, consists of nichrome elements embedded in a ceramic material. The function of the heat barrier is to reduce heat losses from the furnace and thereby reduce the total heat required in the experiment. By operating the furnace with the heat barrier it is possible to self-heat waste-clay-flux mixtures containing fewer curies of fission products. When using the heat barrier in a self-heating experiment, the thermal controls are preset to allow small thermal gradients from the furnace to the surrounding insulation, thus assuring internal heat solely attributable to the decaying fission products. The heat barrier can also be used to provide limited amounts of external heating in experiments where the wastes will not self-sinter.

Off-Gas Cleaning System

Gases, vapors, and particulates released during mixing and firing are exhausted to the off-gas system shown in Figure 4. The system, located in two shielded concrete tanks 7 feet in diameter by 10 feet deep, consists of four units: (1) a condensate storage vessel; (2) a packed tower containing a solution of 20% NaOH; (3) a recirculating caustic scrubber; and (4) a multi-bed filter. Since the off-gas collection and decontamination devices are

Fig. 2. Adiabatic Furnace, Showing Inner Container and Cover.

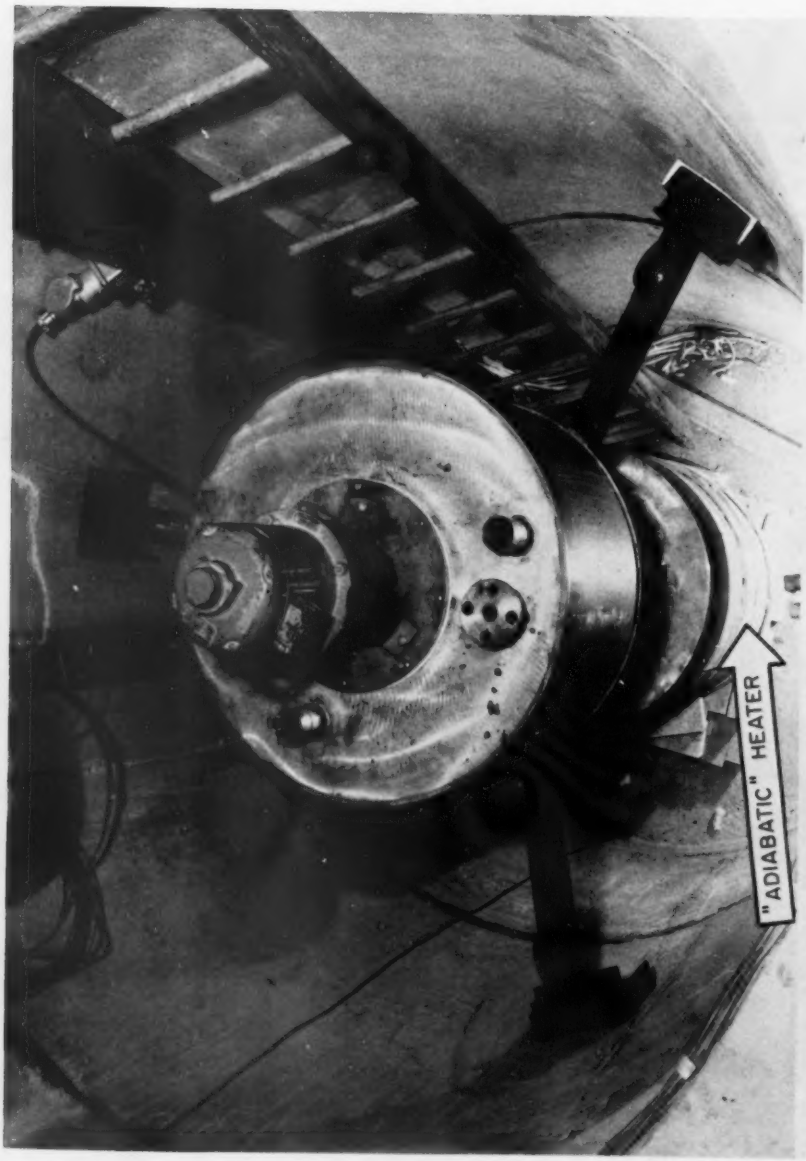

Fig. 3. Top View of Assembled Furnace and Mixer, Showing "Adiabatic" Heater.

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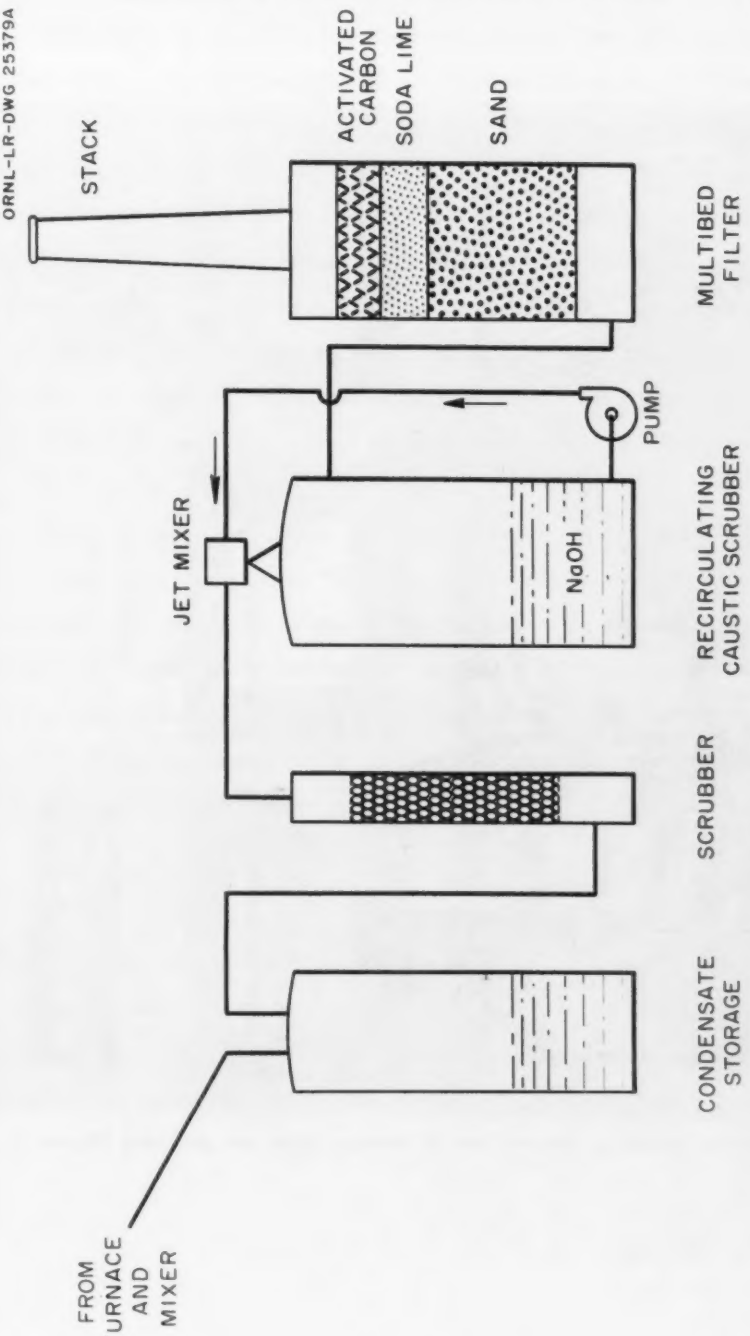

Fig. 4. Schematic Diagram of Off-Gas System.

Fig. 3. Top View of Assembled Furnace and Mixer, Showing "Adiabatic" Heater.

located about 50 feet from the furnace, condensible vapor released during the dewatering stage will condense in the off-gas line and drain to the condensate vessel. From this vessel the gases will enter the packed tower, which removes CO_2 and some of the fission product gases. Gases leaving the packed tower will pass through the jet mixer in continuous contact with a 20% caustic solution until all chemically reactive gases and fission products have been removed. Any remaining gas (mainly the noble fission product gases) will pass through a multibed filter, composed of successive layers of sand, soda lime, and activated carbon, before release to the atmosphere. The vacuum required to exhaust these gases from the furnace and mixer is provided by the jet mixer. To evaluate the performance of each unit or combination of units, bypass lines around all vessels except the condensate vessel are installed, thus it is possible to operate any number of these units in any sequence. Isokinetic sampling points, both fore and aft of each unit, are included to determine the efficiency of each unit or combination of units. Provisions are made to fractionate the sampled gas stream into condensible gases, non-condensible gases, and particulates. The central sampling station is located in a shielded cubicle atop the two shielded concrete tanks containing the off-gas units.

Thermal Measurements and Power Controls

All recording instruments and power controls are located in one building. An adjoining structure houses the devices for power regulation. Thermocouples for measuring and controlling temperature are located in the ground, the liner annulus, the furnace insulation, and inside the furnace. Continuous recording devices for 36 thermocouples are provided (Figure 5).

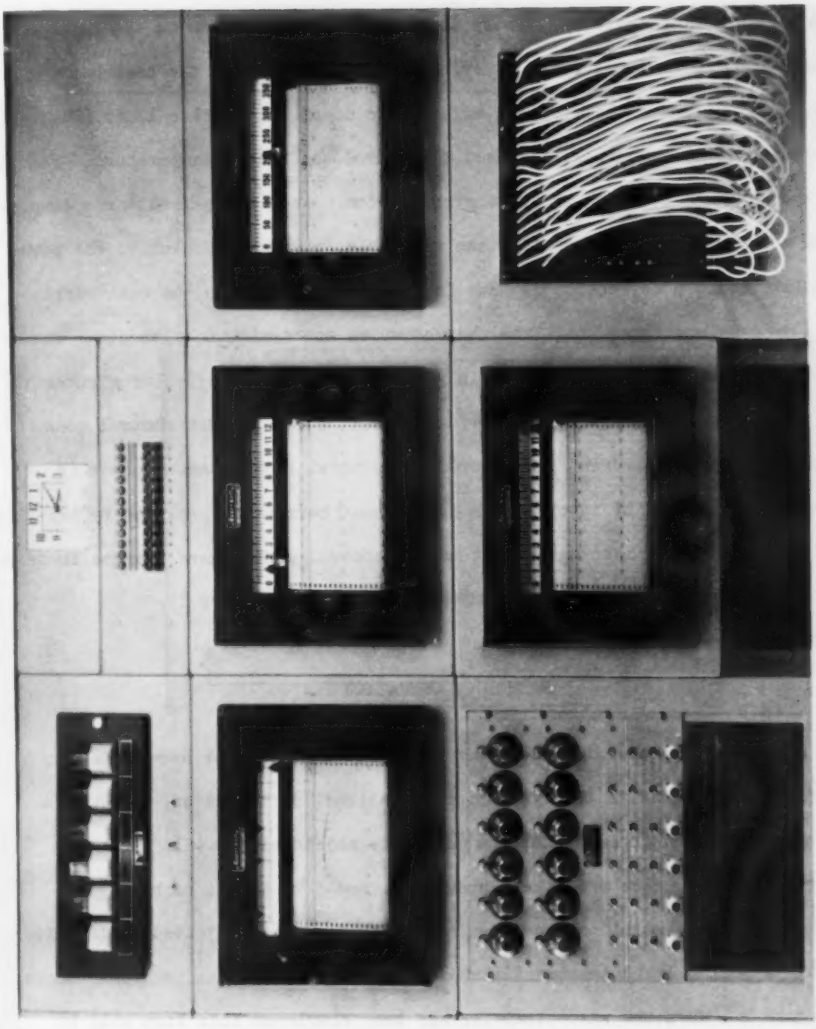

Fig. 5. Temperature Measurement and Power Control Panel.

Additional thermocouples are located in the heat barrier to control the surface temperature of the heating elements. There are six separate control zones in the heat barrier: two on the furnace bottom, three on the furnace sides, and one on the furnace cover. Each thermocouple on the heat barrier has a corresponding thermocouple adjacent to it on the furnace liner (Figure 6). These paired thermocouples are used to control the heater temperature. Figure 7 is a diagrammatic sketch of the control system. When the paired thermocouples show a thermal gradient greater than allowable, a signal is sent to the power regulators and a correction is made in the power applied to the heat barrier. This process continues until the thermocouples are again in balance. A complete scanning of the six control zones is made automatically every four minutes.

Power measurement is provided by wattmeters on each control zone. In addition, an instrument for recording the total instantaneous power is provided. Flexibility in operation of the heat barrier is provided by bringing the leads from each heating element to the control panel where changes in heater units can be made by using wire jumpers and switches (Figure 8).

METHOD OF OPERATION

Prior to charging the mixer with waste, it will be necessary to check all components in the system for operational difficulties. When this has been completed three of the four concrete shield blocks will be put in place over the furnace and mixer, providing from 3 to 5 feet of biological shielding. Waste solution from a suitable carrier will be jetted to the mixer through a portable and temporarily shielded line connected to the valve shown in Figure 1. When the waste transfer has been completed this line will be

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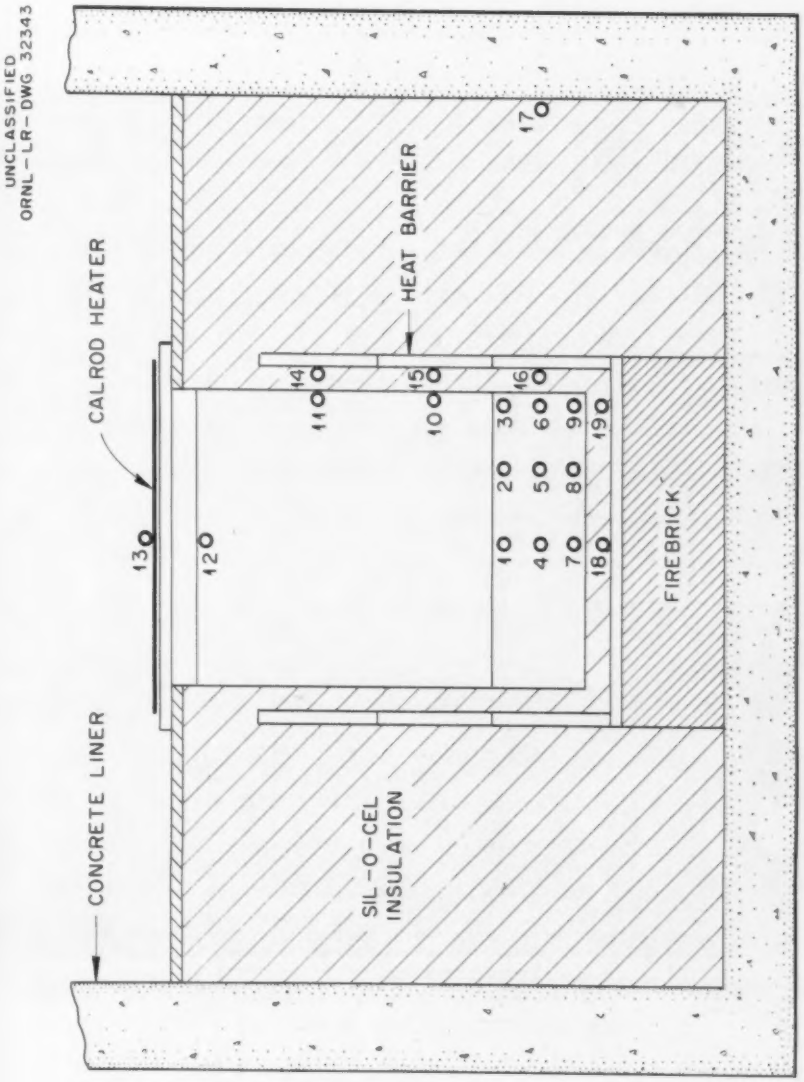

Fig. 6. Location of Furnace Thermocouples.

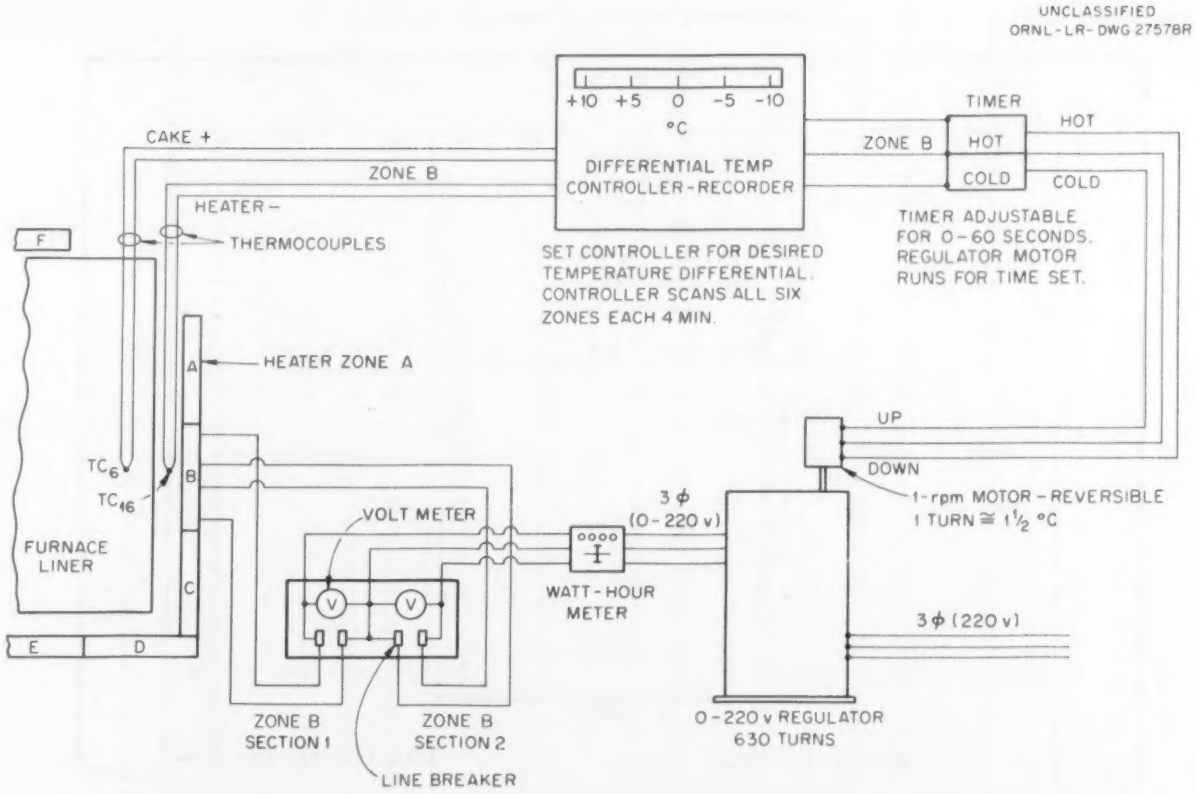

Fig. 7. Adiabatic Heater Control for Heater Zone B.

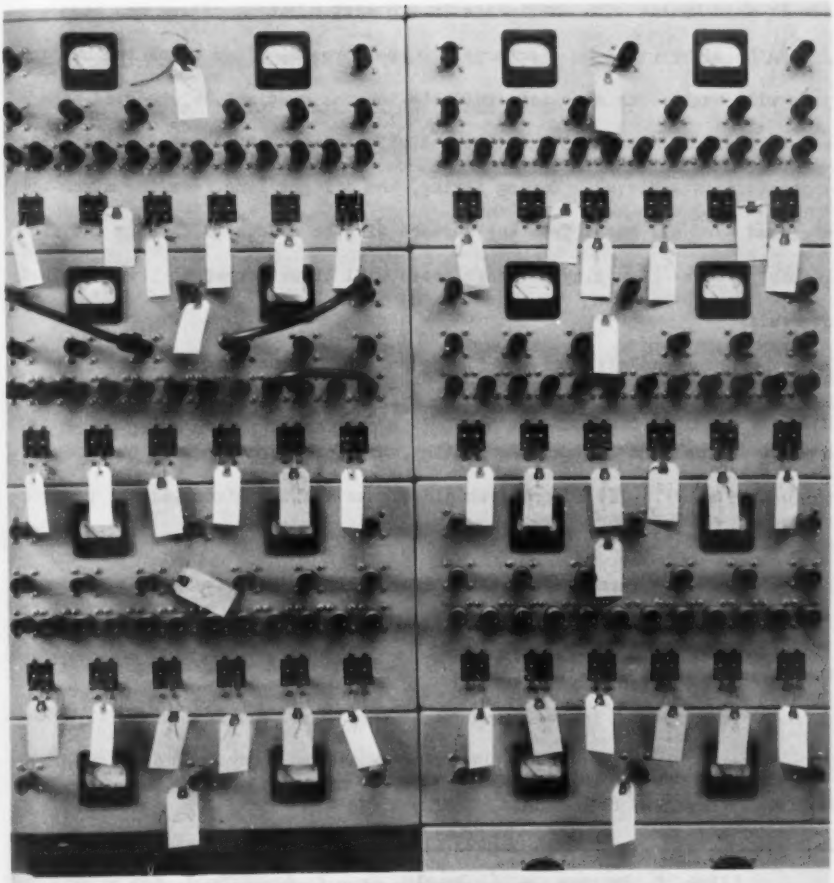

Fig. 8. Power Connections to Heat Barrier, Showing Jumper Connections.

Fig. 7. Adiabatic Heater Control for Heater Zone B.

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disconnected, and the last shield block will be positioned. The solid ingredients of the waste-clay-flux mixture will then be added to the mixer by means of the solids feeder. During the mixing process the mixer will be vented to the off-gas system for the evacuation of CO_2 produced by the reaction of acid wastes with carbonates in the system. When mixing has been completed the slurry will be discharged to the furnace, after which the heating cycle will begin. As soon as a definite thermal gradient is observed, the heat barrier will start to function and will operate until the final fixation temperature is achieved. During firing, the off-gas line from the mixer will be closed, and all gases from the furnace will be evacuated through the air-cleaning system. Sampling of these gases will be performed at predetermined intervals.

Upon completion of the firing period the material in the furnace will cool slowly to the ambient temperature. After a period of time sufficient to allow thermal and radioactive cooling, the fired product will be removed for further study. The concrete shield blocks and the mixer will be removed to permit sampling of the final sinter with the aid of a core-drilling assembly. Finally, the inner furnace assembly (inner cylinder and cover) will be removed and loaded into a shielded container for transfer to the "hot" cell, where final leach testing will take place.

Under normal operation it is expected that the facility will be used for a number of experiments. A preliminary hazards study for the first high-level experiment has been made.[1]

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(1) L. C. Emerson, W. J. Boegly, Jr., F. A. Gifford, and W. de Laguna, "Preliminary Report of the Hazards Associated with the Adiabatic Self-Sintering Experiment," unpublished report, 1958.

ACKNOWLEDGMENT

The authors would like to express their appreciation to C. W. Hancher and W. R. Winsbro of the Chemical Technology Division, ORNL, for their assistance in the design of this facility.

DISPOSAL OF HIGH-LEVEL WASTES BY FIXATION IN FUSED CERAMICS

By

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The Atomic Energy Commission Research Project -- Contract No. AT(30-1)-621 -- at the Sedgwick Laboratories of Sanitary Science at M. I. T. has been directed toward the problem of the disposal of high-level wastes since 1956. These are the wastes which will originate in large quantities and with high concentrations of radioactivity from the operation of atomic power plants. Present methods of tank storage of fission product wastes from the reprocessing of irradiated reactor fuel elements will not suffice for the future when volumes of these high-level wastes will reach hundreds of millions of gallons.

The research team at M. I. T. has directed its attack on a method for the concentration of these reactor fuel reprocessing wastes to reduce their volumes and convert them to solids. Archeological findings indicate that glasses are a very stable form of materials over the centuries. Since concentrated fission products must be stored under controlled conditions for centuries it was deemed advisable to attempt to fix the fission products in glasses which could then be buried in permanent "cemeteries" under continuous surveillance by monitoring systems.

Two fundamental types of high-level radioactive wastes are currently being produced in the United States in the reactor fuel processing plants. Wastes of the first type contain the extracted fission products in essentially pure nitric acid; the second type contain high concentrations of aluminum salts. The research at M. I. T. is concerned with the second type and involves the conversion of acid aluminum nitrate fission product wastes to relatively

insoluble, high melting point glasses which can be stored safely in the ground. These glasses are vitreous ceramic systems which can serve as retention matrices for aluminum-bearing wastes. A similar approach is being employed for reactor fuel reprocessing waste solutions containing zirconium and stainless steel which are used for cladding modern reactor fuel elements. The incorporation of these solutions in ceramic systems appears to offer a number of advantages: 1) aluminum oxide, a necessary constituent of many hard glasses, can be formed readily by thermal decomposition of the aluminum nitrate already present in the waste; 2) hard glasses are generally designed to resist chemical action and weathering; and, as mentioned previously, archeological evidence supports the claim for long-term stability of these materials.

The general class of compositions selected for study are those loosely described as "porcelain glazes" (1). These are primarily members of the $CaO:Al_2O_3:SiO_2$ system, modified in some cases by the addition of varying amounts of alkali metal or other alkaline earth oxides as fluxes. These fusions are among the most refractory of all glazes, but possess properties which compensate for the somewhat greater expense and difficulty of forming them. They are highly insoluble in water and in most alkalies and acids, and are mechanically strong and hard. These properties, plus the ability to incorporate small fractions of impurities without marked effect, recommend this class of compositions for the function of fixing a relatively small mass of fission products with a large mass of aluminum oxide in a very stable matrix.

Glass can be defined as a supercooled liquid, a state which possesses the tangible characteristic of a solid, but which exhibits none of the discontinuous changes characteristic of the transition from the liquid to the crystalline state. Materials used in the formation of glasses have been

classified chemically in three groups: 1) bases, including the alkalies and alkaline earths, as network modifiers; 2) intermediates, which include the amphoteric oxides of which alumina and ferric oxide are examples, and with which boric anhydride is usually grouped by ceramicists; and 3) acids, the network formers represented by silica and zirconia. Each of these groups plays a significant role in the development of the properties of the final glass product.

Silica is the predominating compound in almost all glasses because of its ability to combine with many other oxides to form an extensive variety of complex silicates. These silicates can be made to exist at normal temperatures as extremely viscous undercooled solutions. Increasing the silica content of a glass will generally raise the melting point, decrease the fluidity and increase the hardness and resistance of the glass to chemical attack. Although the alkalies are strong fluxes, increasing the fluidity of the melt, their use tends to lessen the resistance of the fused product to weathering and chemical attack. The alkaline earths are represented most frequently in glasses by calcium, whose presence is said to increase hardness, resistance to the solvent action of water and tensile strength. Increasing the alumina content of a glass increases its melting point, hardness and resistance to chemical attack. Boron is used in glass (among other reasons) to increase fusibility and fluidity and to hinder the crystallization of other compounds, thus inhibiting devitrification.

Research on this project by Goldman (2), Servizi (3)(4), Flood (5), Tebbutt (6), Burns (7), Daniels (8) and Lauderdale (9) was directed toward determining the feasibility of incorporating aluminum-bearing fission product wastes into fused ceramic glazes of the general type - Al_2O_3:CaO:SiO_2:B_2O_3 flux. The resultant fusions are in the broad classification of "porcelain glazes". Various modifications of alkali metal or other alkaline earth oxides,

or fluxes, were investigated, with a view toward obtaining the lowest possible melting point and the greatest retention of fission products. Two series of glazes were prepared to investigate the effect of some composition parameters on the degree of retention of fission products in the fused material. Cesium-137 was the tracer used in the initial studies. Other isotopes were used later. The first series of experiments developed the effects of lime and sodium tetraborate flux concentration; the second compared the effects of sodium tetraborate and boric anhydride on the stability of the glazes and the effect of increasing alumina concentration on retention characteristics and fusibility of the product.

The products were prepared by mixing with the proper volume of synthetic waste (1.6 $\underline{M}$ $Al(NO_3)_3$ + 0.2 $\underline{M}$ HNO_3 + fission product carriers + Cs-137 tracer), a slurry of the lime, borax or boric anhydride, and sand. The mixture was evaporated to dryness, and heating continued to drive off nitrates. The resulting white friable solid was broken up and powdered, and transferred to a Battersea fire clay crucible. This was preheated to about 800° C. to insure complete denitration, and then fired. After the glaze had matured for times ranging from about 3 to 24 hours, the melt was poured into a graphite slab, fractured and the pieces remelted in graphite molds for about 5 minutes to form single pellets.

The pellets weighed between 2 and 4 1/2 grams each, were fairly smooth and transparent with a slight greenish tint. Each pellet was weighed, the specific gravity determined, and the dimensions measured to estimate surface area, which ranged from about 5 to 8 cm^2 per pellet as calculated. Those pellets with sufficiently low activities (less than about 200,000 counts per minute) were counted directly in a gamma scintillation well counter. The activity per gram of the more active pellets (up to about 6×10^6 counts per minute) was determined by measuring the activity of small portions of the unused melt. Any loss of cesium activity during firing was determined by comparing the measured activity per gram with that calculated for each mixture from the initial waste activity.

Since it is hoped that the concentrated fission products in ceramic glazes can be disposed of by burial on waste lands, it is necessary to evaluate the stability of the ceramic in the presence of ground waters and any other weathering conditions which might exist in nature. Therefore, an attempt was made in the laboratory to simulate the natural conditions which would exist in the burial grounds.

Methods used by various laboratories to determine the degree of retention of fission products in different solid media vary considerably. The reagents which have been used in leaching studies have included simulated sea water, warm water and dilute nitric acid. Preliminary work at this laboratory brought out two significant facts: 1) cesium was the fission product most readily leached from the glaze types under study; and, 2) erratic and statistically questionable results were obtained when the product was leached in distilled water at room temperature. A study was, therefore, initiated to determine the effect of temperature on the rate of leaching. The results show that temperature has an exponential effect, with the leaching rate at boiling temperature between 100 and 1000 times greater than at room temperature.

Because of the increased sensitivity afforded by the higher temperature and the ease of maintaining the temperature constant, it was decided to carry out all of the leaching tests in boiling water. In addition, it was felt that testing under these conditions would be a closer approximation to actual field conditions, since it is reasonable to assume that any ground water coming into contact with stored ceramics will probably be at a high temperature due to the heat generated by fission product decay. To leach-test a glaze, each of five samples of one glaze composition is suspended by a 30 gauge Chromel wire in 100 ml. of boiling distilled water in a reflux apparatus. Fig. 1 shows a battery of leaching vessels.

The results obtained in this study indicate the general effectiveness of this class of materials as matrices for fission product fixation. The range of fractional leaching rates observed extended from about 10^{-5} to 2×10^{-4} per cm^2-week in distilled water at 100° C. A maximum volume reduction factor of about seven was obtained from the original 1.6 M aluminum nitrate waste solution. An increased alumina content has a beneficial effect on the retention characteristics of the mixtures tested, although the effect is minimal above an alumina/lime ratio of about 0.6 mole/mole. An increased alumina content also raises the product melting point and appears to reduce losses of cesium by volatilization. The presence of sodium oxide in the mixtures tested increases the leaching rates by factors ranging from about two to ten. The fused product can be formed from the liquid aluminum-bearing waste, or from alumina currently being produced by calcination of these wastes.

The storage of fused products will result in their absorption of high radiation doses from the contained fission products. An integrated heat generation was estimated on the basis of published data on heat production rates of irradiated fuel elements. This energy liberation was converted to radiation absorbed dose (rad) for glazes, assuming a waste volume of 5

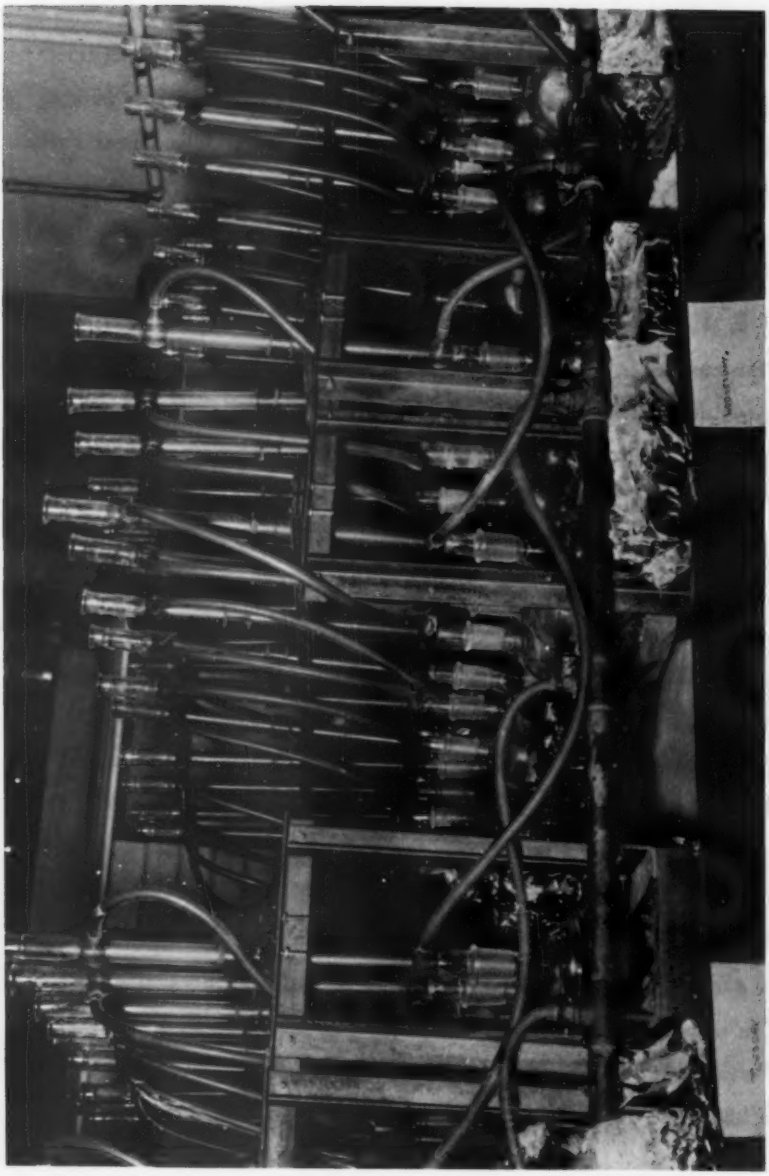

FIGURE 1. Leaching apparatus for testing fission product retention characteristics of ceramic glazes.

gallons per gram of U-235 fissioned and other glaze characteristics, (density, volume reduction factor). The calculations indicate that if 120 days cooling prior to reprocessing is assumed, a dose of 10^8 rads would be absorbed by the fused product during the first year of storage, 10^9 rads after 50 years, and a value at 500 years of about 1.2×10^9 rads. Tebbutt (10) exposed samples of each glaze to the M. I. T. cobalt-60 gamma source for times sufficient to provide doses in the order of 10^7 and 10^8 roentgens. This was the maximum attainable range in reasonable exposure times with this source. The exposed pellets were markedly discolored by the irradiation, but no other physical effects were noted. These samples were subjected to the leaching test with an unirradiated control for each glaze. The data obtained to date indicate no significant differences in the leaching rates between controls and those pellets irradiated up to 10^8 r.

Arrangements were then made to utilize a spent fuel storage facility at the National Reactor Test Station to irradiate samples to doses of 10^9 r. Results of these tests led Tebbutt (10) to the following conclusions: (a) the approximate radiation dose received by an infinite block of glass containing the radioactive waste from the processing of U-235-Al fuel elements irradiated for 85 days with 30 per cent burn-up would be 1.3×10^9 rads; (b) gamma irradiation at levels up to and including 10^9 r has no detrimental effect on the fixative properties of the glasses tested. In many cases significant reductions in the leaching rate of Cs^{137} were noted after irradiation; (c) no relation could be established between glass composition and the effect of irradiation on leaching; (d) the rate of removal of Cs^{137} from the glass was found to be directly related to the solubility of the glass, indicating that selective leaching of the cesium did not take place, and (e) cesium is volatile when mixed with glass constituents and fired at temperatures in the range 1400-1500° C. The loss of cesium appears to be reduced by the addition of a larger

characteristics of ceramic glazes.

amount of Al_2O_3 but this is probably only a physical effect since increasing the Al_2O_3 content leads to a higher viscosity at the firing temperature used.

Daniels (8) studied the volatile loss of cesium during calcination and fusion at temperatures up to 1300° C. Preliminary studies showed that cesium losses ranged from 0 to 70 per cent of the initial waste activity. Due to this wide range, an investigation was initiated to develop a method for a more quantitative determination of cesium volatilization during firing and the influence of such factors as glaze composition and firing conditions. Initial experimentation was performed on the pure compound $CsNO_3$ with Cs-137 added as the tracer isotope. The nitrate form was chosen because of its probable presence in the fuel reprocessing waste. Following the satisfactory collection of volatilized cesium from $CsNO_3$, several glaze compositions were investigated. The procedure for volatility measurement consisted of: 1) placing a weighed and counted sample of $CsNO_3$ or glaze mix in a tared nickel combustion boat in the center of a combustion tube furnace; 2) passing a stream of air or pre-purified nitrogen through the tube to sweep the volatile decomposition products past a cold finger and through a sorption train; 3) changing the stainless steel cold fingers and off-gas lines and replacing the Millipore filters during the run; 4) leaching the cold fingers and all surfaces in contact with the gas stream with 1N HCl followed by distilled water; and 5) evaporating and counting the leachate in a Geiger-Mueller counter. Total volatile losses due to firing were determined by counting the sample in the combustion boat before and after the run in a low geometry position on top of a gamma scintillation well counter.

As a result of this investigation a method was developed for the determination of cesium losses during the pyrolysis of $CsNO_3$ and from ceramic glazes using a high efficiency stainless steel cold finger to collect the volatilized activity. The following information was derived from this study: (a) volatilization of cesium from $CsNO_3$ begins in the molten state above 450° C. The

largest fraction of cesium is evolved below 650° C. and volatilization is completed by 900° C.; (b) the maximum evolution of cesium from glaze mixes occurs at 1000° C. as compared to 650° C. for $CsNO_3$, and (c) retention of cesium increases with increasing alumina concentration, in the alumina, lime, sand and boric anhydride mixtures fired to 1000° C. in this investigation.

Ruthenium is one of the problem elements among fission products. Very little is known about the chemical characteristics of this element, the isotopes of which have mass numbers between 101 and 108. From the radiological health standpoint, ruthenium-103, with a half-life of forty days, and ruthenium-106, with a half-life of one year, are the significant radioisotopes. Difficulties with ruthenium occur from the first stage in the fuel reprocessing operation and continue throughout each step into the waste treatment stage. Since it volatilizes readily, it would be advisable to remove ruthenium from the liquid radioactive wastes before fusion into a ceramic glaze. The studies of Lauderdale (9) were particularly concerned with the factors affecting the removal of Ru (III) and RuNO (III) from liquid radioactive wastes by contacting the solution with metallic iron, and with the mechanisms by which the removal of ruthenium is accomplished. The environmental factors considered include pH, the concentration of various ions normally found in dilute and high level wastes, and the concentration of the radioactive ruthenium. It was found that trivalent ruthenium can be effectively removed from liquids using metallic iron through the mechanism of electrochemical reduction. Nitrosyl ruthenium (III) can be removed either by electrochemical reduction or, in some cases, through mutual coagulation of colloidal ruthenium and ferrous iron. Calcium, chloride, and sodium ions appeared to have no effect on the removal of ruthenium. Nitrate ion was found to interfere under conditions in which it acts as an oxidant, and aluminum

ion interfered under the conditions in which it precipitated as the hydroxide.

Concentration of other radioisotopes, particularly strontium-89, yttrium-91, cesium-137 and cerium-144 was attempted from the concentrated fission products in a dibasic aluminum nitrate solution. Work was not done on the fission products themselves, but rather on a simulated waste as previously described. This waste permitted valid conclusions to be obtained in a research laboratory designed for tracer studies.

Using montmorillonite clay as an exchange medium, work was conducted by Egan (10) with batch and continuous processes. The fill-and-draw process consisted of mixing different amounts of wastes and clays for various times and determining the reduction in activity in the supernatant. The continuous flow process consisted of passing the dibasic aluminum waste through a clay-sand mixture and determining the reduction in activity in the effluent. Standard type G-M beta counters were used in determining the activity and removal of activity through use of this process. The results show that for the conditions under which the tests were run, strontium is the least removable radioisotope, while cesium is adsorbed quite readily on the clay. Cerium and yttrium are adsorbed to a lesser degree than cesium but more so than strontium. In addition, the batch type process showed sufficiently better removals than the continuous flow process which might indicate that this method would be the most feasible scheme of treatment. It was apparent from this work that it would be more desirable to direct the activities of the research personnel to studies on the fixation of fission products in ceramic media.

Burns (7) investigated the feasibility of incorporating an aluminum-zirconium bearing waste into a ceramic solid of glaze-type composition. The glazes were formed into small test pellets of the general composition - Al_2O_3:CaO:SiO_2:B_2O_3 together with varying amounts of zirconyl nitrate (up

to 0.62 moles per mole of alumina). To determine the amount of activity leached from the cesium-137 tagged test pellets, a weekly analysis of the leaching water was made. The leachate was evaporated and its contents counted on a Geiger-Mueller counter to determine the removed activity. In this manner calculations could be made to determine the retention properties of the various glazes. Results from these tests indicate that it is definitely feasible to incorporate a zirconium-aluminum bearing waste into a ceramic body. In addition to this it was noted that as the zirconium content increased a better retention of the cesium tracer was evident.

Work on this project, together with the extensive literature surveys on wastes disposal, has led to a portion of a textbook. Eliassen and Lauderdale (11) have written the chapter entitled "Liquid and Solid Waste Disposal" in the forthcoming book edited by Blatz and titled "Handbook of Radiation Hygiene". This chapter presents some of the research findings in a manner useful to designing engineers and operators of nuclear facilities.

On the basis of this extensive series of studies on the fixation of fission products in ceramic media, Eliassen (12) published the following summary of the situation to date: "The industry is faced with the prospect of disposing of about one ton of fission products per day by 1980. Even after a year of storage for the decay of relatively short-lived products, the total activity of these fission products will exceed 100 billion curies/yr. Of course, fusion power may develop sufficiently to radically reduce the waste. Nobody would dream of trying to dispose of this quantity of wastes to natural water courses where the tolerance value for mixed radioisotopes is 10^{-7} μc/ml., or 420 c. per cubic mile of water. Almost equally absurd would be storage for this amount of radioactivity as the accumulated solutions of high-level processing wastes would exceed 200-million gallons by 1980.

At M.I.T. we have found that the retention of fission products in ceramic

glazes shows real promise. Ceramic glazes and glassy materials have demonstrated exceptional stability over centuries of attack by water and weather. It is feasible to incorporate aluminum-bearing fission-product wastes into ceramic glazes, thus getting rid of the tremendous quantities of aluminum nitrate resulting from the chemical processing, as well as fission products in one operation. The systems that have shown the greatest promise to date are (1) $CaO-Al_2O_3-B_2O_3-SiO_2$ and (2) $Na_2O-CaO-Al_2O_3-B_2O_3-SiO_2$. Much more work remains to be done, not only on aluminum, but also on zirconium and other metals that may be alloyed with reactor fuel material and that will be present in the wastes from fuel-processing plants. The important point is that work is being pursued actively by many research and operating organizations and that progress to date has been most promising. Advancing technology in the processing of wastes containing fission products will most assuredly keep abreast of, and probably forge ahead of, the prophesied rate of growth of nuclear power plants."

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THE DISPOSAL OF FISSION PRODUCTS IN GLASS*

L.C. Watson, R.W. Durham, W.E. Erlebach and H.K. Rae

INTRODUCTION

1.1 With increasing use of nuclear reactors for power production, it will be necessary to devise safe and economical methods for disposing of fission products. While storage of intensely radioactive solutions in underground tanks as presently practiced is reasonably safe barring catastrophic failure of the tank, a method by which the fission products could be stored as a solid from which they could not escape would be much superior. The general requirements of a suitable solid are:

(a) Resistance to removal of fission products by water

(b) Low cost for conversion of liquid to solid by a simple process

(c) Easy and cheap storage with minimum future control

(d) Resistance to damage by radiation, or by chemical changes associated with the decaying nuclides.

1.2 Studies in these laboratories indicate that glass can meet these requirements satisfactorily. Confirmation can however be obtained only by operation of a large-scale processing and storage system.

1.3 Canadian proposals for power reactors are based on the use of unenriched, unalloyed fuel irradiated to a high burnup, and at present the fuel which is considered most suitable is uranium dioxide sheathed in Zircaloy-II. It is with a fuel of this nature in mind that the disposal system for fission products is being developed. It is assumed that the fission products would appear in a waste solution from a plant extracting plutonium and recovering uranium.

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* Contribution from Atomic Energy of Canada Limited, Chalk River, Ontario

The power reactor fuel and the waste solution derived from it are described in Table I.

Table I

Characteristics of Power Reactor Fuel and Waste Solutions

Fuel	- natural uranium dioxide
Sheathing Material	- Zircaloy-II
Burnup	- 10,000 MWD/tonne
Irradiation time	- 1,000 days
Desheathing	- mechanical, or chemical if sheathing material is kept separate
Fuel processing	- by a process using tri-butyl phosphate (1)
Waste solution volume (after evaporation)	- 0.25 litre/kg of uranium processed
Waste concentration	- 8 Molar HNO_3
	35 g of fission products/litre
	10 g of corrosion products/litre
	5×10^3 curies/litre after 180 days out of the reactor
Heat production due to fission products	- 15 watts/litre at 180 days.

This waste solution is considered most desirable as feed to the glass conversion process since it contains a minimum of inactive elements. However, wastes containing sheathing materials (e.g., Al or Zr) in solution can also be treated, although with consequent increases in the volume of glass per gram of fission products.

SELECTION OF GLASS

2.1 Any raw material used for production of the glass must be reasonably uniform chemically, so that the properties of the product can be predicted and relied upon. Such uniformity can, of course, be obtained by mixing suitable chemical compounds. However, nepheline syenite, a material commonly used in the glass and pottery industry is mined from an unusually uniform deposit in Ontario and is available cheaply as a finely ground powder. A typical analysis (2) of this material follows:

SiO_2	60.24 per cent	CaO	0.15 per cent
Al_2O_3	24.05	Fe_2O_3	0.06
Na_2O	10.03	MgO	0.02
K_2O	5.01	TiO_2	0.002

Loss on ignition 0.046 per cent

2.2 Nepheline syenite melts at about 1250°C and on cooling forms a glass. The viscosity of the unfluxed melt at 1350°C is high and as a result this glass is bubbly, exposing more surface than is desirable. Furthermore, it will not readily incorporate certain elements which may be expected in fission-product solutions. The addition of a flux circumvents both these difficulties and calcium hydroxide, which is inexpensive, has been found to be satisfactory.

2.3 When the acid solution is mixed with nepheline syenite, the nepheline is attacked and produces a gel of silica. This has proved to be advantageous since such a gelled mix may be dried rapidly without serious entrainment of particulate material during the removal of water, nitric acid, oxides of nitrogen and other volatile components. If the nepheline syenite and calcium hydroxide are blended as powders prior to mixing with the acid solution, the gel forms regardless of calcium hydroxide content up to the equivalent of 35 weight per cent oxide in the glass. If the calcium hydroxide used is in excess of that required to neutralize the acid the time required for gelling is extended.

2.4 These properties combined with the high resistance of the glass to attack by water led to the choice of nepheline syenite fluxed with calcium oxide as a glass meeting the requirements for disposal of fission products.

THE RELEASE OF FISSION PRODUCTS BY THE ACTION OF WATER

3.1 The effectiveness of a disposal medium for retaining fission products is dependent on the resistance of the medium to the corrosive action of water. In general, the addition of alumina to glass increases its melting temperature and decreases its solubility. The constitution of nepheline syenite is such that if lime is added as flux the glass formed approximates a lime glass to which alumina has been added. Glass of this composition should have, and in fact is found to have, a high resistance to attack by water.

3.2 The mechanism of the solution of glass in water is complex. The glass does not dissolve per se but the more soluble elements are leached out leaving a surface film of hydrated silica. However, it is convenient to consider that the surface of the glass is uniformly attacked despite these differential effects. Thus, the measurement of the quantity of a radioactive nuclide leached into water from a glass specimen is most useful when converted to the equivalent glass corrosion rate expressed as grams of glass dissolved per cm^2 per day. Any sorption effects are neglected.

3.3 Glass samples for leaching tests were prepared by three methods. In Method I, Cs-137 in nitric acid solution was added to the mixture of nepheline syenite and flux in a recrystallized alumina dish. The mix was heated to 1350°C and kept at this temperature for one hour. After cooling, the dish contained about 100 mc of Cs-137 in 4 g of glass with a nominal surface area of 16 cm^2. Since the glass could not be removed from the dish, the resulting test specimen consisted of dish plus glass. Method II involved neutron irradiation of a cast pellet of glass which contained one per cent by weight of the inactive parent element. Small fibres of the glass were irradiated in the same irradiation container for the determination of specific activity. Glass samples prepared by Method III were mixed and melted as in Method I, but were cast into pellets after melting.

3.4 The dissolution rates of several glasses were measured by immersing a sample of each in a known volume of distilled water contained in a polyethylene bag, allowing it to stand for a day and then sampling the water for counting (3). After sampling, the leach water and bag were discarded and the glass sample was placed with fresh water in a new bag. The dissolution rate in $g/cm^2/day$ equivalent to the activity found in the water was calculated from a knowledge of the approximate surface area exposed and the specific activity of the glass.

3.5 When any sample of active glass was tested by the above technique, the activity leached per day dropped sharply over the first few days, followed by a gradual decrease over the period of the tests. In Table II are listed the dissolution rates of various glasses. These rates were measured after several months of testing, that is, at a time when the leaching rate was changing very slowly. The results plotted in Figure 1. were obtained over a period of six

months from one sample in Experiment A (Table II).

Table II

Dissolution Rates of Glasses in Distilled Water at 25°C

Experiment	Glass Composition by weight	Method of Sample Preparation	Tracer nuclide	Dissolution Rate $g/cm^2/day$
A	15% CaO; 85% N.S.[(a)]	I	Cs-137	5×10^{-8}
B	35% CaO; 65% N.S.	I	Cs-137	5×10^{-7}
C	20% CaO; 20% BaO; 60% N.S.	I	Cs-137	5×10^{-7}
D	20% CaO; 20% BaO; 60% N.S.	II	Cs-134	5×10^{-8}
E	20% CaO; 20% BaO; 60% N.S.	II	Eu-152	1×10^{-8}
F	65% PbO; 10% B_2O_3; 25% SiO_2	III	Cs-137	1×10^{-5}
G	55% PbO; 15% B_2O_3; 25% SiO_2; 5% compounds of Al, Na, K, F	III	Cs-137	5×10^{-6}

[(a)] N.S. = nepheline syenite

3.6 The distilled water used in these experiments was made in an aluminum still and stored in a polyethylene rather than a glass container to avoid contamination. One of the samples in Experiment A, however, was leached in ground water (pH 8.2) but the dissolution rate was found to be identical with that of a similar sample in distilled water (pH 6.8)

3.7 A comparison of Experiments C and D shows that the dissolution rate was an order of magnitude higher when the glass was prepared by Method I rather than by Method II. This anomaly would occur if tracer Cs-137 concentrated in the surface of the glass during processing. However, no macroscopic variation in the specific activity of the glass was found by breaking up the glass and counting samples from the surface and the interior.

3.8 The high dissolution rates observed for the lead borosilicate glazes, Experiments F and G, make them decidedly inferior to the nepheline syenite glass for waste-disposal purposes, despite the advantages of their low melting points.

3.9 The results of Experiments D and E show that the leaching rate of the trivalent europium was five times less than that of the monovalent cesium. It would appear therefore that europium is either less easily separated from the glass or that, once in solution, it is more readily sorbed by the silica film formed on the surface of the glass.

3.10 When glass samples were left in water for five to six weeks without being disturbed, the average daily dissolution rates were found to be about three times higher than those measured at daily intervals. (Figure 1). When the water was again changed daily the dissolution rates dropped back rapidly to the previous low value. These results suggest that accumulation of alkali in the leach water may increase the leaching rate.

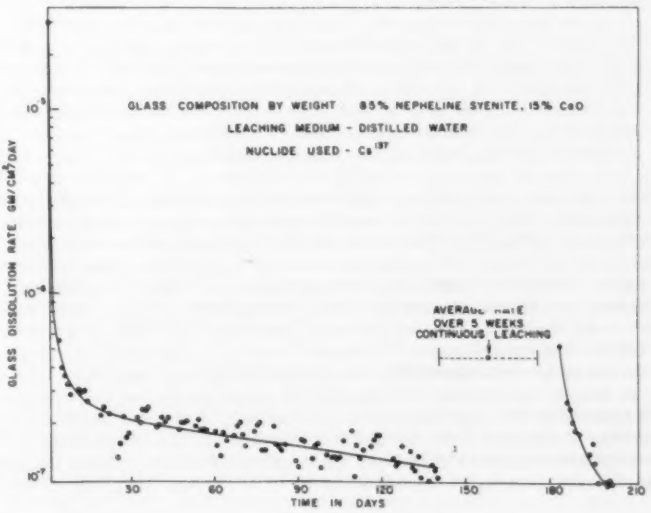

Figure 1. Variation of Glass Dissolution Rate With Time

3.11 Present results indicate that a glass composed of 15 per cent CaO and 85 per cent nepheline syenite offers the best compromise between high resistance to attack by water, which requires low flux concentrations, and low melt viscosity, which requires high flux concentrations. This glass, although relatively viscous at 1350°C , readily incorporates five weight per cent of metal ions, and, as can be seen from Table II, has a very low dissolution rate.

VOLATILIZATION OF RUTHENIUM AND CESIUM

4.1 A troublesome feature of the incorporation of fission products in glass is the volatilization of two abundant fission products, cesium and ruthenium. Both these elements have forms which are volatile over a substantial fraction of the temperature range through which the mixes of fission products, nitric acid and glass-forming materials must be heated. The development work on this aspect of the nepheline syenite process has had a threefold purpose: to determine a mechanism by which the volatile form of the element is produced; to modify the process so that little of the volatile material is formed; and to devise means of trapping efficiently the radioactive elements evolved from the mixes.

4.2 The mechanism for the volatilization of ruthenium appears to depend on the acidity of the mix and on the temperatures to which the mix is heated (4). The losses observed during a number of experiments can be conveniently classified into those occurring above or below 900°C and those occurring from acid or basic mixes. Basic mixes result from the addition of calcium hydroxide in excess of that required to neutralize the acid present.

4.3 The ruthenium in solution is present largely as nitrato complexes of nitrosyl ruthenium with one, two or three nitrato groups associated with each ruthenium nitrosyl group (5). When an acid mix is heated the nitric acid concentration increases and the complexes tend to become completely nitrated. Further heating leads to oxidation and decomposition of these complexes to form the volatile ruthenium tetroxide. If the initial acid concentration is high enough nearly all the ruthenium can be removed from a mix heated to 400°C. When the initial acid concentration is decreased to less than 0.5N, the extent of nitrato complexing is reduced, and the retention of ruthenium in mixes heated to 900°C is sharply increased. Moreover, by adding sufficient basic flux to the nepheline syenite one can prepare an alkaline mix which retains over 99 per cent of the ruthenium. It is thought that nitrosyl nitrato complexes in such a mix are hydrolyzed to a nitrosyl hydroxide of ruthenium which decomposes to the non-volatile ruthenium dioxide.

4.4 At temperatures above 900°C a different mechanism is responsible for the volatilization of ruthenium. Evidence indicates that this is the air oxidation of ruthenium dioxide to the volatile tetroxide. The advantages of

preparing a basic mix to retain ruthenium are entirely lost when the mix is heated from 900°C to 1250°C. Up to 70 per cent of the ruthenium present after denitration can be removed from a mix in this temperature range. The fact that this loss can be reduced to 13 per cent by firing the mix in an atmosphere of carbon dioxide supports the hypothesis that air oxidation is responsible for the ruthenium volatilization.

4.5 Although the examination of cesium losses from mixes is still in a preliminary stage, cesium loss appears to be independent of the acidity of the mix. Volatilization commences between 400 and 500°C near the indicated decomposition temperature of cesium nitrate. The cesium oxides produced during denitration are unstable and probably dissociate immediately to produce cesium. A decrease in the rate of cesium loss at temperatures in the vicinity of 900°C indicates that the rate of a second reaction between cesium and a constituent of the mix becomes greater than the rate of diffusion of cesium from the mix and that the product of this second reaction is thermally stable. A large variation in measured cesium losses indicates that the rates of these opposing processes, diffusion or oxidation, are probably sensitive to small changes in such variables as mix temperature, porosity, and geometry. Under different conditions of mix preparation and heating rates cesium losses as low as one per cent and as high as 30 per cent have been measured.

4.6 Some progress has been made in the development of a method for collecting the ruthenium volatilized from a mix. A stainless-steel surface placed in the gases evolved from a heated basic mix collected over 98 per cent of the volatilized ruthenium. Under comparable heating conditions such a trap removed only half of the ruthenium evolved from an acid mix. The deposit showed no tendency to revolatilize when the steel was heated to a maximum temperature of 1000°C. Considerable improvement in the collection of ruthenium from acid mixes was obtained when the gases were passed through a heated bed of firebrick granules coated with oxides of iron. Collecting efficiencies of more than 98 per cent have been obtained with a 10 cm bed at a temperature of 800°C, and a total gas-flow rate of 30 cm/sec. Furthermore, no significant loss of ruthenium from the coated granules occurred at temperatures up to 1350°C.

4.7 The retention of ruthenium at high temperatures on coated firebrick and on stainless steel can be directly attributed to the presence of the oxides of iron. This has been shown in two ways: first, the volatile ruthenium deposited on a platinum or a vitreous-silica surface from a denitrating or denitrated mix can be revolatilized by heating in air at 500°C; second, uncoated firebrick retains a negligible amount of ruthenium.

4.8 Under the investigated conditions of drying, denitrating and fusing mixes in air, the overall losses of fission-product ruthenium and cesium by

volatilization are not significantly different for acid or alkaline mixes. Under these circumstances the calcium oxide content of the glass should be primarily regulated to produce a leach-resistant glass. There is, however, room for optimism in the development of a method for reducing this volatilization. It should not be difficult to eliminate the air oxidation of ruthenium at high temperature by firing the mix in a suitable atmosphere. Furthermore, by reducing the rate of diffusion of cesium from the mix and increasing the rate of formation of a thermally stable form, some improvement in the cesium retention can be foreseen.

5 PROCESS FOR INCORPORATING FISSION PRODUCTS INTO GLASS

5.1 A flowsheet by which fission products may be incorporated into glass blocks is presented in Figure 2.

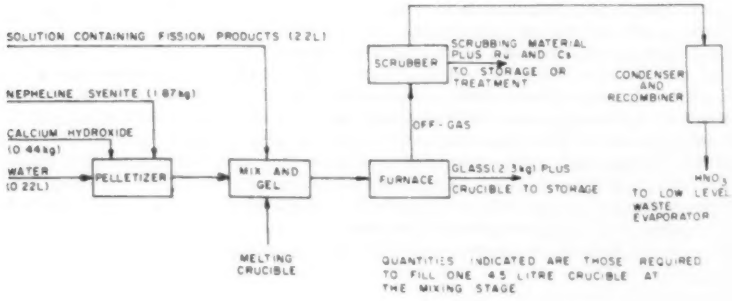

Figure 2. Incorporation of Fission Products into Glass

A mixture of calcium hydroxide and nepheline syenite is pelletized by tumbling with 10 weight per cent water. Pellets between 0.83 and 3.3 mm are selected by screening; fines and oversize pellets are rejected. Four hundred grams of pellets followed by 400 ml of solution are loaded alternately into the melting crucible until it is full. The capacity of the crucible is 4.5 litres but it will contain only 2.2 litres of solution mixed in this way. This technique avoids a mechanical mixing operation. A mix made with 8N acid solution and

pellets containing 15 weight per cent lime gels in less than 10 minutes. If the lime is increased to 30 weight per cent, gelling takes several hours.

5.2 The melting operation is straightforward and may be done in any high-temperature (1350°C) furnace from which the off-gases can be directed to appropriate scrubbers. Refractories must be selected with due regard to their reactivity toward the volatilized Ru and Cs. To remove volatilized Ru and Cs from the gas streams a scrubber charged with iron oxide supported on granular firebrick and operated at 400 to 800°C is provided. The off-gases then pass to appropriate equipment for recovery of the acid which is further treated as a low-level waste.

5.3 Equipment has been constructed to incorporate several thousand curies of aged fission products into glass according to this flowsheet. The solutions which are currently available at Chalk River contain 25 to 50 curies of mixed fission products per litre, and the operation is batchwise on the scale of about 100 curies. Figure 3 is a photograph of a high-temperature furnace used in this work.

5.4 The primary purpose of this program is to provide a moderately large source of fission products in glass with which data obtained in the laboratory, particularly with respect to stability to water, may be confirmed. Specifically, an attempt will be made to assess the possibility that such glass may be buried safely in the ground in selected areas. Other aims of the work are to assess the general operating features of the process, particularly the firing operation and the gas-cleaning equipment, in order to assemble data applicable to the design and operation of a large-scale plant.

PERMANENT STORAGE OF GLASS CONTAINING FISSION PRODUCTS

6.0.1 The principal reason for converting to a solid the bulk of the fission products resulting from the processing of irradiated nuclear fuels is to circumvent the hazards associated with their storage in a liquid form. Selection of a method for permanent storage of this solid requires a careful study of all the factors involved. The following discussion applies in detail to the storage of nepheline syenite glass containing fission products, but in general to the storage of any alternative solid material.

6.0.2 The four major considerations in choosing a method and site of permanent storage are:

(a) assurance against dispersal of activity

(b) thermal effects due to heat generated by fission-product decay

(c) physical size requirements and availability of site, and

(d) costs.

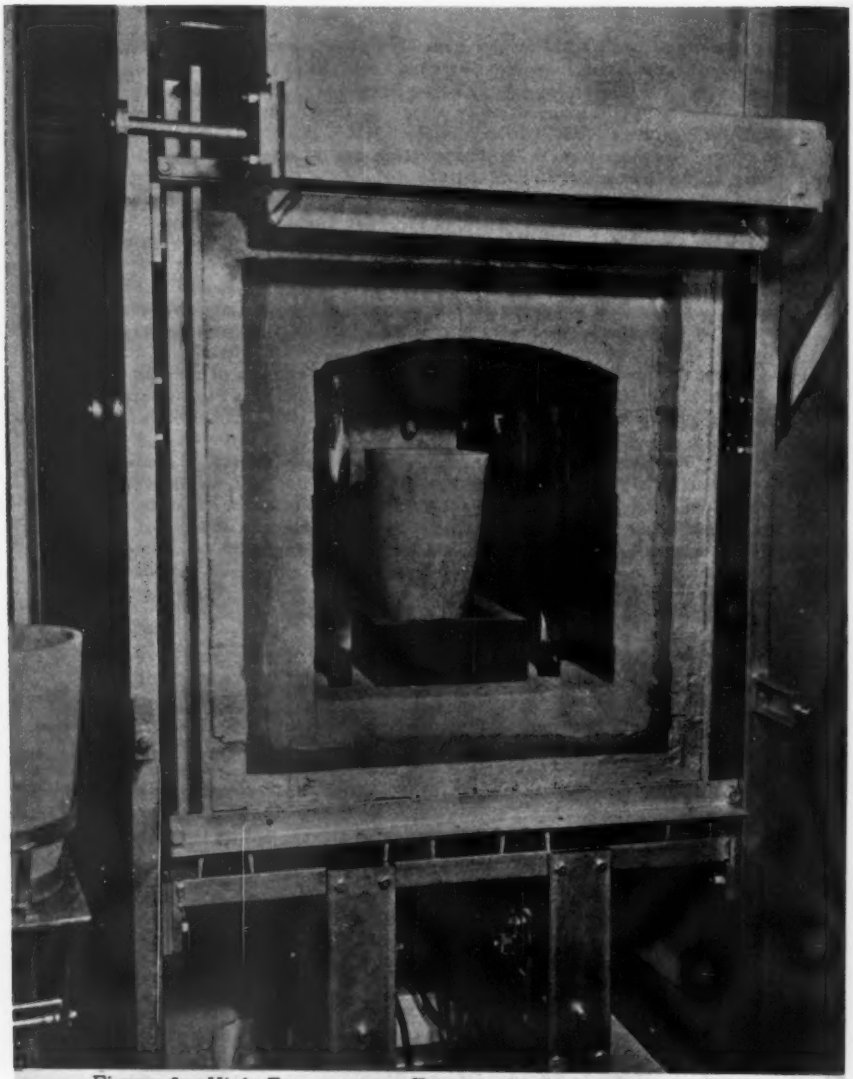

Figure 3. High-Temperature Furnace for Incorporation of Fission Products into Glass

In a recent report by Watson et al (6) these factors were assessed and a number of possible storage methods were compared. A reactor capacity of 4000 MW thermal, which is expected to be reached in Canada by about 1973 (7) was selected as the basis for calculations and discussion.

6.1 Dispersal of Activity by Leaching

6.1.1 It has been shown earlier that the rate of leaching of activity from nepheline syenite glass by immersion in water is very low; a conservative figure corresponds to the dissolution of 3×10^{-7} g of glass per cm^2 per day. It is assumed that this apparent dissolution rate applies to the leaching of all nuclides. The case of a waste-disposal program for a series of reactors having a total capacity of 4000 thermal megawatts has been specifically considered. If the fission products produced by these reactors were incorporated into glass leaching at 3×10^{-7} g/cm^2/day, the cumulative figures for radionuclides leached from the glass would be as set out in Table III.

Table III

Cumulative Leached Activity from Nepheline Syenite Glass Containing Fission Products Produced Continuously at a Constant Rate

Nuclide [a]	Cumulative Leached Material [b] Curies		
	1 year	5 years	25 years
Sr^{90}	15.2	380	6460
Ru^{106}	30.5	399	400
Cs^{137}	22.6	566	9670
Ce^{144}	73.3	537	540

(a) daughter activities are not included

(b) method of calculation is outlined in Appendix A

The fission products are assumed to result from the irradiation of natural uranium to 10,000 MWD per tonne in 1000 days and to be incorporated into the glass 120 days after removal from the reactor; the glass is assumed to have a surface to volume ratio of 0.2 cm^2/cm^3. For comparison with the figures shown in this Table, the quantity of Sr-90 present in the glass in the storage area after five years is 13×10^6 curies.

6.1.2 Despite the excellent resistance to leaching exhibited by the glass, the activities set free by continuous leaching as shown in Table III are large considering the relatively small capacity of the series of reactors assumed. The values of the parameters used in this example represent a practical case, and although longer cooling times prior to storage could be assumed, they would not reduce significantly the cumulative quantity of activity released by leaching.

6.1.3 If the production of the glass is stopped after a period of ten years, the cumulative quantity of Sr-90 and Cs-137 leached from the glass will each remain in excess of 1000 curies for the conditions in Table III for a period of more than 100 years.

6.1.4 For the case of ground water contacting the glass, estimates have been made (6) of the maximum leaching rate that may be allowed if the concentration of Sr-90 is not to exceed 8×10^{-10} curies per litre, i.e. the maximum permissible concentration for drinking water (8). Using a flow rate for ground water of one volume of water per volume of glass per day, a specific surface area of 0.2 cm^2 per cm^3, and a Sr-90 concentration in the glass of 6×10^{-4} g per g, the maximum allowable leaching rate was calculated as 5×10^{-11} g of glass per cm^2 per day. This is lower by a factor of about 1000 than that measured in the laboratory.

6.1.5 These considerations indicate that the glass should be stored in a dry location and in such a way as to prevent any accidental contact with water. However, complete water exclusion over a storage time of several hundred years cannot be guaranteed; thus the low rate of leaching from nepheline syenite glass remains a desirable characteristic.

6.1.6 An alternative approach is the development of a suitable coating or glaze to reduce the leaching rate by a large factor, along with a storage method in which the glass is normally dry but where no great effort is made to exclude water at all times.

6.2 Thermal Effects Due to Heat Generation

6.2.1 The total quantity of heat generated in the storage area by radioactive decay of the fission products can be quite large. When wastes are being

accumulated at a constant rate from reactors having a total capacity of 4000 thermal megawatts (Table III), the heat-generation rate is estimated to be 1000 kW after ten years and ultimately 2300 kW at steady state. The fission products are assumed to be incorporated into the glass 120 days after removal from the reactor; increasing this cooling time would result in only a small decrease in the heat-generation rate at the end of several years of operation.

If the glass is stored in vault-like structures it will probably be necessary to limit the wall temperature to relatively low values and air cooling by either forced draught or natural convection will be required. If the glass is buried in the ground, high temperatures may be permissible and cooling by conduction to the ground surface will be adequate.

Table IV

Temperature of Nepheline Syenite Glass Containing Fission Products Buried in Dry Sand

Glass shape	- 15 cm diameter sphere
Depth of burial	- 300 cm
Conductivity of sand	- 3.3×10^{-3} joules/sec/°C/cm
Conductivity of glass	- 13×10^{-3} joules/sec/°C/cm
Ground surface temperature	- 25°C
Specific heat-generation rate in glass	- 35 watts/litre [a]

Method of Estimation	Temperature of Glass Surface	Temperature of Glass Centre
(1) As a single isolated sphere (no interaction with other spheres)	200°C	250°C
(2) As an infinite horizontal cylinder of diameter 12.5 cm, equivalent to a single row of spheres buried in a trench	960°C	990°C
(3) As an infinite horizontal slab of thickness 0.3 cm, equivalent to a regular lattice of spheres each with 0.6 m^2 of ground surface	970°C	970°C

[a] Glass made from waste solution of the type described in Table I, 180 days after removal from the reactor

Table V

Methods of Permanent Storage of Glass Containing Fission Products

Method	Release of Activity by Leaching	Thermal Considerations	Comments
1. Burial in contact with ground water	high	small blocks to avoid excessive temperatures	poor method
2. Burial in dry ground	low - some intermittent leaching possible	small blocks to avoid excessive temperatures	promising method - allowable glass temperatures and effects of intermittent leaching require study
3. Storage in caverns			
(a) natural caverns, mineshafts, etc.	high, unless sealed	cooling required	difficult to seal
(b) specially constructed in unfaulted rock	nil	cooling required	promising method - study of possible locations and of thermal effects required
4. Storage in concrete vaults	nil	cooling required	promising method - least difficult to develop
5. Storage in drilled holes	low if hole cased	diameter limited	too costly
6. Burial in permafrost	unknown	unknown	limited accessibility and high costs are disadvantages
7. Disposal in the ocean	high	none	depends on the acceptance of uncontrolled contamination

6.2.3 The specific heat-generation rate in the glass at the time it is produced will be in the range of 3 to 70 watts per litre depending on the age of the fission products and the type of fuel being processed. Estimates have been made (6) of the temperatures which would be reached in glass of various shapes buried in the ground. An example is given in Table IV where it is shown that, to avoid excessive temperatures, the glass must be buried as relatively small blocks and so arranged as to restrict thermal interaction. This results in large areas of ground being required initially for burial, although more glass can be added to a burial site as the older material decays so that reasonably efficient utilization of area can be obtained over a long period.

6.2.4 If the glass is immersed in a large body of water its surface temperature will always be relatively low and, even at high specific rates of heat generation excessive centre temperatures will not be reached unless the diameter of the glass block approaches one metre.

6.3 Physical Size Requirements

6.3.1 If the wastes from reactors having a total capacity of 4000 thermal megawatts are stored as nepheline syenite glass, the rate of production will be in the range from 50 to 700 litres of glass per day depending on the types of fuel being used and the details of the chemical processes involved. Allowing suitable utilization factors, the annual storage volume will vary from 40 to 1300 m^3. For burial in the ground the area required annually will be of the order of 10^4 m^2 with some re-use in the future possible.

6.4 Comparison of Storage Methods

6.4.1 The methods of storing nepheline syenite glass which have been considered (6) are listed in Table V. The most promising methods are those in which dry ground conditions prevail, namely, burial in dry ground, storage in specially constructed caverns, and storage in concrete vaults. If the burial site is not in an arid region water may be excluded by capping with asphalt.

6.4.2 It is not possible at present to choose a storage method which shows an over-all advantage in terms of economics and safety. Some of the problems requiring further study in the case of burial in dry ground or storage in rock caverns are mentioned in Table V.

ESTIMATED COSTS

7.1 Cost of Producing Glass Containing Fission Products

7.1.1 Rae and Hollies (9) have made a preliminary cost estimate for the nepheline syenite process based on a simple flowsheet in which the concentrated

waste solution is mixed with nepheline syenite in small crucibles (4.5 litres total capacity) which are heated through the stages of drying, denitration, fusion and annealing in a continuously-operating rotating-hearth furnace. These crucibles are then transferred to a suitable storage area. Each one originally contains 2.2 litres of solution which is transformed to 2.3 kg of glass. The operations are carried out in a large concrete cell provided with windows, a crane and heavy-duty manipulators. Provision is made for remote maintenance and 30 per cent excess capacity is assumed to allow adequate maintenance time for the two furnaces.

Table VI

Estimated Cost of Producing Nepheline Syenite Glass Containing Fission Products

Plant Capacity - 850 litres of concentrated fission-product solution per day
Capital Cost - $1,000,000 (a)
Operating Time - 330 days per year

	Dollars per year	Dollars per litre
Capital charges	200,000	
Labour (2 men per shift)	34,000	
Overheads	157,000	
Maintenance	50,000	
Raw Materials (except crucibles)	20,000	
Total A	461,000	1.6
Crucibles (390 per day)	232,000	
Total B	693,000	2.5

Case A assumes that the crucibles can be re-used a number of times.

In Case B the glass is stored in the crucibles. At present it is not possible to separate the glass from the crucible without destroying the latter.

(a) This includes all equipment, building, cell, ventilation, etc. but assumes construction at a site which is already developed with adequate services available.

7.1.2 A plant capacity of 850 litres of concentrated waste solution per day was studied. The evaporation of the wastes to a minimum volume was assumed to be carried out in the chemical processing plant (1), and the cost was not included in this estimate.

7.1.3 The estimated cost for producing glass containing fission products was 1.6 to 2.5 dollars per litre of solution as shown in Table VI.

7.1.4 For the type of fuel described in Table I, the above cost will contribute only about 0.01 mills per kWh to the cost of power. This contribution will be larger with solutions containing lower concentrations of fission products, but it is unlikely to exceed 0.05 mills per kWh.

7.2 Storage Costs

7.2.1. Some approximate calculations have been made(6) to indicate the order of magnitude of storage costs. The construction of a concrete storage vault of 1400 m^3 capacity was estimated to cost 17 cents per litre. The glass, when stored in its crucible, might occupy 20 per cent of the total volume so that the storage cost would be about one dollar per litre of glass. It is expected that the construction of rock caverns should be cheaper than buried concrete vaults. The cost of burial in the ground depends on the rate of heat generation in the glass and on the allowable temperature. For the glass considered in Table IV buried in trenches the cost of excavation and backfill was estimated to be 13 cents per litre of glass. No attempt has been made to estimate handling costs, routine monitoring and inspection costs, or other possible operating charges.

7.2.2 For an irradiation of 10,000 MWD per tonne with processing to yield 0.12 litres of glass per kilogram of uranium, a storage cost of five dollars per litre of glass would contribute about 0.01 mills per kWh to the cost of power. Considering the estimates given above, the cost of permanent storage is not likely to exceed 0.01 mills per kWh.

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APPENDIX A

The following method was used in calculating the cumulative quantities of leached fission products appearing in Table III.

$$c = \frac{Sya\phi P}{\rho\lambda}\left\{1 - (1 + \lambda T)e^{-\lambda T}\right\}$$

where c is the cumulative amount of any nuclide in curies leached from the glass after T days of continuous production.

S is the leaching rate = 3×10^{-7} g of glass/cm^2/day

a is the specific activity of the nuclide in curies per gram

ϕ is the surface to volume ratio of the glass - 0.2 cm^2/cm^3

P is the total power capacity of the reactors (= 4000 MW thermal), assumed to produce 4000 g of total fission products per day

ρ is the density of the glass = 2 g/cm^3

λ is the decay constant of the nuclide in days^{-1}

y is the concentration of the nuclide in g/g of total fission products at the time the glass is produced

For natural uranium at 10,000 MWD/tonne in a typical heavy-water-moderated reactor

$$y = \left[\frac{0.55\ Y_{235}}{235} + \frac{0.45\ Y_{239}}{239}\right]\left[\frac{A\,(1 - e^{-\lambda D})e^{-\lambda T_c}}{100\lambda D}\right]^{(a)}$$

where Y_{235} and Y_{239} are the atom per cent yields of the nuclide in the fission of U-235 and Pu-239, respectively

A is the atomic weight of the nuclide
D is the irradiation time = 1000 days
T_c is the cooling time = 120 days

(a) constant flux assumed. neutron capture neglected.

THE USE OF CERAMIC SPONGES FOR ULTIMATE DISPOSAL OF RADIOACTIVE WASTES

By
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Los Alamos has worked on fixation of radioactive waste solution in clays for the past six or seven years. Because the present methods of waste disposal at Los Alamos are deemed satisfactory the work on clay fixation was not stressed from an operational standpoint. It was determined, however, that almost any type of clay, when mixed with radioactive waste solution, dried, and subsequently fired to about 1200°C. will retain the radionuclides. The resultant brick is very resistant to leaching with the common leaching agents which could be expected. Although most clays demonstrate a remarkable ability to retain the nuclides, the nature of the final product, i.e., solid form, strength, lack of dust, etc., depends upon the characteristics of the clay. The various types of clays and their mixtures that have been tried are too numerous to mention here.

The greatest difficulty involved in the use of clay fixation is in the mechanical method of mixing the clay and waste. The various methods of handling radioactive wastes in this manner are covered very adequately in a "Report of Working Meeting on Fixation of Radioactivity in Stable Solid Media" which was held at the Johns Hopkins University June 19-21, 1957 (TID-7550).

Since that meeting a new method of fixation in clays has been proposed. It was suggested by representatives of

Coors Porcelain Company, Golden, Colorado, in March 1958 that liquid radioactive wastes might be soaked up in ceramic sponges, dried and fired thus permanently fixing the radionuclides in the ceramic. A similar, independent, investigation was reported by Robinson (1) in April 1958.

Basically, the process involves the preparation of a highly porous clay body which is fired to about 800°C. This body is then soaked in the liquid waste, dried, resoaked and the cycle repeated as many as three, four or five times. The clay body is finally fired at a temperature of about 1300°C. to permanently fix the radionuclide.

The clay sponges are made in a variety of ways. These include burn-out materials such as naphthalene, wood, cork or other organic materials; using gas bubbles with foaming agents; and using highly absorptive materials such as calcined clays, diatomaceous earth, etc. The initial or "bisque" firing temperatures and cycles are major factors in production of maximum porosity. The optimum pore sizes for effective absorption will be determined by experimentation. The final product must necessarily be strong and dust-free.

In preliminary work done at Los Alamos with sponges furnished by Coors it has been found that some of the ceramics will absorb a Purex waste to about 50 percent of their weight. The amount of waste absorbed by the sponge is essentially the same for each of the first four soaking and drying cycles.

After four cycles are completed, subsequent soaking and drying absorb considerably less waste than the previous cycles. In several determinations a sponge has absorbed a Purex waste up to 200 percent of its weight after four cycles. Additional cycles increase this percentage but at a diminishing rate. In some tests, simulated Purex wastes were spiked with Sr^{90} and Cs^{137}, absorbed in ceramic sponges and fired to 1300°C. The subsequent leaching by tap water and distilled water showed essentially no radioactivity in the lixivium.

The work to date has not advanced to a point where cost estimates can be made. However, it appears that the costs will probably not exceed those of storage of liquid wastes and the storage of wastes in a solid form surely presents a much less hazardous manner of containment of these wastes than storage of the liquids in tanks. It appears that the radionuclides are very firmly fixed to the ceramic and no great care need be taken in storage other than providing shielding for radiation protection.

One other advantage to this method of disposal lies in the area of valuable use of the radioactivity in the wastes. The clay sponges can be formed in almost any shape which makes it possible to contain the radiation in some physical form in which the gamma and beta rays may be useful. Because the radioactivity is so well contained this might mean beneficial use of the heat of decay or radiation for

sterilization of foods, etc. The one disadvantage lies in the fact that the volume occupied by the sponge-radionuclides will be somewhat larger than the original waste volume and highly concentrated sources will not be possible.

The above method of disposal of liquid radioactive wastes has shown considerable promise, hence plans are underway for a full scale investigation of the process. One of the major problems to overcome will be the treatment of the off-gases associated with the drying and final firing of the ceramics.

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FLUIDIZED-BED CONVERSION OF FUEL PROCESSING WASTES TO SOLIDS FOR DISPOSAL

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A recent AEC report[1] indicates that the present inventory of high level liquid wastes in tankage in the United States is greater than 60 million gallons, and it is estimated that by June, 1959 this volume will have been increased an additional 10 million gallons. The practice of storing these liquid wastes in underground tanks was begun during the war as an expedient stopgap. The system has worked quite well, but the tanks can hardly be considered an adequate storage measure for a time sufficient to decay the radioactivity to an acceptable degree of innocuousness. From a safety standpoint it would be highly desirable to convert these wastes to storable solids. A number of methods for the conversion of liquid wastes to solids have been considered,[2,3,4,5,6,7,8,9] including simple concentration until the wastes solidify on cooling, addition of portland cement, adsorption on clays and clay flux mixes, fused salt calcination, and incorporation into silicate glass matrices. This paper describes another possible method for conversion of liquid wastes to solids, which makes use of the process of fluid-bed calcination.

Development of Fluid-Bed Aluminum Nitrate Calcination Process

At the Idaho Chemical Processing Plant, in which highly enriched uranium fuels are processed for recovery of fissionable material, a principal high activity waste results from treatment of aluminum alloy fuels of the MTR type. This is an acid aluminum nitrate solution which is presently stored in cooled underground stainless steel tanks. In the interests of economy and long-term safety, consideration has been given to methods of converting this waste to a stable solid. The aim has been to develop an economical and mechanically simple process which provides a significant reduction in the availability of fission products from the stored material to the environment as compared to liquid storage. At the Argonne National Laboratory,

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the development of a fluidized-bed process for the conversion of a chemically similar uranyl nitrate solution to uranium oxide has been carried out.[10] Because of the simplicity of the conversion operation, similar studies were undertaken at ANL on conversion of aluminum nitrate to alumina. Advantages conceived of were the absence of moving parts in the conversion device, applicability to continuous processing, ease of transporting the product to storage, and utilization of alumina coming from the fuel alloy as a fission product retaining medium. Following promising initial pilot plant tests, both laboratory and pilot plant work were undertaken by the Atomic Energy Division of Phillips Petroleum Company at the National Reactor Testing Station to develop a complete process for this waste, and studies have continued at ANL with calcination of wastes at up to one per cent of full activity level.

Process Description

The principle used in the fluid-bed calcination of waste is illustrated in a simplified manner in Fig. 1. The calciner consists of a cylindrical vessel containing a bed of granular oxide, consisting of the product obtained from the evaporation and calcination of the waste solution. For aluminum nitrate-bearing wastes, such as those produced in recovering enriched uranium from aluminum alloy fuels and in the Redox solvent extraction process, the granular bed would consist of aluminum oxide together with other minor constituents including fission product oxides. For wastes of low solid content, such as those produced in the Purex process, the bed could consist of almost any inexpensive, inert, porous solid.

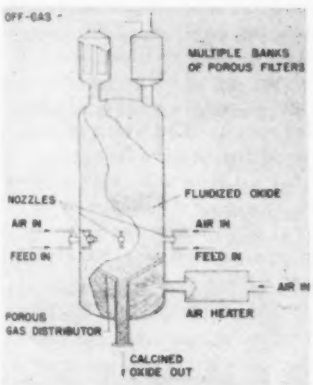

Fig. 1. Fluidized Bed Calciner

The bed is supported on a perforated or porous plate, which serves as a gas distributor. Preheated air is passed upward through the plate to fluidize the bed and agitate the solids, the entire dense mass behaving much like a vigorously boiling liquid. Heat is supplied to the solid particles through the vessel wall and/or through heat transfer tubes within the bed itself. The bed temperature is maintained in the range of 400 to 500°C.

The waste solution is injected into the bed through several pneumatic spray nozzles spaced around the perifery of the calciner in a single horizontal plane. The liquid droplets, upon contacting the hot solid particles, are flash dried and calcined to oxides. The product solid is deposited on the particles in the bed, and continuously withdrawn from the bottom or from an

overflow pipe. Sufficient attrition occurs to produce new particle nuclei and to counteract any tendency for the bed particles to continually increase in size.

The off-gas rising from the fluidized-bed consists of air mixed with decomposition gases, including water vapor and oxides of nitrogen. Entrained radioactive oxide dust contained in the gas stream can be removed either by the use of sintered metal filters as shown in Fig. 1 or by liquid scrubbers. In either case, absolute (AEC-type) filters will be used to back-up the primary dust separators. A certain amount of fission-product ruthenium is volatilized into the off-gases during the calcination process, and removal of this volatilized ruthenium requires additional equipment. For this purpose the use of either a partial condenser or an absorber containing silica gel or stainless steel wool is being considered.

PILOT-PLANT STUDIES (NON-RADIOACTIVE)

Initial experimentation to determine the feasibility of converting aluminum nitrate solutions to granular solids was conducted in a 3-inch diameter stainless steel unit with a 4-inch glass pipe disengaging section. Later work was done in larger units. Four pilot-scale fluid-bed calciners have been operated, two by Argonne National Laboratory and two by Phillips Petroleum Company. All of these units were similar in design principle to the illustrative unit shown in Fig. 1, but contained slight differences. The units in use at ANL are both 6 in. in diameter one unit being used for non-radioactive studies and the other, a shielded unit, being used for studies with radioactive feed. Heat is supplied to the ANL units by electrical elements attached to the wall outside the vessels. Porous stainless steel plates are used for fluidizing gas distribution. The off-gases evolved are passed alternately through one of two banks of sintered stainless steel filters to remove particles. These filters are blown back to remove dust particles when not in service, and the dust drops back into the fluid-bed.

Two pilot-scale fluidized bed calciners have been operated by Phillips Petroleum Company at the National Reactor Testing Station.[11] One unit is 6 in. in diameter to process up to 7 liters of aluminum nitrate solution per hour, while the other vessel has a cross-sectional area of four square feet and is designed to process up to 100 liters/hour. Heat is supplied to these units through tubular surfaces placed in the bed of solids. The smaller unit contains electrical elements while the larger one may be heated either electrically, or by circulation of NaK alloy heated in an oil furnace through heat transfer tubes in the bed, or by passage of combustion gases through tubes in the bed.

Fluidizing air is supplied through bubble-cap plates which support the bed. The granular calcined product overflows continuously into a catch

pot or can be removed intermittently through a bottom draw-off line. Liquid feed is sprayed into the calciner through nozzles located flush with the inside of the vessel wall below the operating level of the fluidized bed but well above the heat exchanger tubes. The nozzles are equipped with cleanout needles.

In the units operated by Phillips, first stage particle removal from the off gas is effected by a scalping cyclone. Separated fines are returned to the fluidized bed by connecting the suction side of an air jet to the solids discharge end of the cyclone. The air jet is motivated by fluidizing air, which returns the solid particles to the bed.

For additional particle removal, the off gas is passed through a venturi scrubber which utilizes a dilute nitric acid scrub solution obtained by condensation from the off gas. Droplets and scrubbed particles are separated from the gas in a second cyclone. The scrub solution is cooled and recycled and a portion is continuously drawn off to compensate for liquid produced by condensation. After passing the off-gas through a cooler and condenser, gases are vented to the atmosphere through an absolute-type filter.

Calciner Performance

Most of the experimental runs have been made using a simulated waste having the following composition range:

Aluminum Nitrate	1.7 - 2.2 M
Nitric Acid	1.0 - 1.3 M
Sodium Nitrate	0.1 - 0.2 M
Mercuric Nitrate	0.005 - 0.01 M

Alumina has been produced at bed temperatures in the ranges 300-550°C, although most of the work has been carried out at 400°C. Starting with a bed composed principally of 20-60 mesh particles, the particle size distribution pattern varies slowly and irregularly, being dependent upon such factors as the relative rates of nucleation, deposition on existing particles, attrition, fines recycle, and product draw-off. A typical particle size distribution is given below:

Per Cent of Particles Retained by						Per Cent Thru
14 mesh	20 mesh	28 mesh	35 mesh	60 mesh	100 mesh	100 mesh
19	28	27	19	6	0.3	0.2

Residual nitrate in the product results principally from the sodium nitrate present and varies with calcination temperature. Typical values at 400°C

ranged from 1.0 to 1.5 milliequivalents per gram of solid. The bulk density of the solid ranged from 48 to 55 lb/cu ft depending primarily upon particle size and size distribution. The volume of the solid was approximately one-eighth that of the feed solution.

Most of these experiments employed air as the fluidizing gas, but the use of superheated steam was also tested in an attempt to reduce the non-condensables in the off-gas and thereby reduce radioactive dust carryover in the gas stream past the condenser. Runs made with steam as the fluidizing medium resulted in very good operation with approximately the same percentage of residual nitrate in the oxide as when air was used.

An attempt was made to use kerosene as a source of heat by direct injection into the fluidized bed (at 500°C), where due to its low ignition temperature the kerosene burned in the fluidizing air. Some heat effect from combustion of the kerosene was noted, but examination of the off-gas disclosed incomplete combustion. The use of indirect heating through the vessel wall or by heat transfer tubes within the bed itself was, therefore, considered to be the most practical method. The sintered porous stainless steel filters were tested and shown to be at least 99.98 per cent efficient for removal of particulate matter formed in the calciner. Of the nozzles tested, the pneumatic atomizing type proved most effective due to its production of very fine droplets and to the absence of caking at the nozzle face encountered with some nozzles. In this type of nozzle the air and liquid are mixed externally after leaving their respective orifices.

Control of Particle Size Distribution

Since fine particles must be removed by off gas treating equipment, it is desirable to minimize their production. Accordingly, control of particle size distribution in the bed, and correspondingly in the product, is most important in calciner operation. Theoretically, for a given bed geometry, the size distribution is a function of the equivalent solid feed rate, the mass of the bed, and the number and size of seed particles introduced per unit of time. Seed particles are produced by attrition in the bed and in the off gas recycle system, and by operation of the feed spray nozzle.

Those feed droplets which do not come in contact with solid particles in the bed will be evaporated and calcined to form new seed particles. Fine atomization will presumably form many particles which may be so small as to pass through the scalping cyclone. It is known that droplets produced by air atomization in a nozzle vary in size as a function of the air-to-liquid volume ratio as well as of the air velocity. High air rates and velocities tend to produce many small droplets. Thus one would expect a small average particle size in a bed fed at a high air-to-feed ratio. This has been noted qualitatively in calciner operation as shown in Table I.

Table I

EFFECT OF NOZZLE AIR RATIO ON PARTICLE SIZE DISTRIBUTION
(For 6-inch calciner at 400°C, aluminum nitrate feed)

Volumes Nozzle Air per Volume Feed	Feed Rate, ml/min	Per Cent of Bed Particle Held On			Per Cent Thru
		20 mesh	35 mesh	60 mesh	60 mesh
231	73	99	1	-	-
294	32	92	6	-	2
441	30	7	69	19	5
396	53	1	68	28	3
455	69	0	50	40	10
665	32	3	62	22	13
580	50	0	26	50	23

Particle Removal by Venturi Scrubber

The primary cyclone on the 6-inch calciner has removed as high as 95% of the solids entering with the off gas. A particle removal efficiency of over 99% has been achieved with the venturi scrubber, to give downstream solids loadings as low as 0.016 mg/cu ft. To get this efficiency required throat velocities of 500 to 1000 ft/sec and scrub rates of 5 to 20 gals/cu ft, and resulted in a pressure drop across the venturi of 15 to 20 in. of Hg. Since it is highly important that even very small proportions of contaminated particles not be discharged to the atmosphere, a multistage cleanup system is probably required for a plant. Detailed requirements must be developed experimentally, but filters, scrubbers, and precipitators in combination may be required.

LABORATORY STUDIES ON FISSION-PRODUCT RUTHENIUM BEHAVIOR

Volatility of Ruthenium Accompanying Aluminum Nitrate Decomposition

Prior to fluid-bed calcining tests with radioactive feed, laboratory studies were carried out to indicate the behavior of ruthenium when aluminum nitrate was decomposed at high temperatures. It was suspected from the known chemistry of ruthenium, that its volatilization during calcination might be encountered.

When a typical waste solution (1.7 M aluminum nitrate, 1.0 M nitric acid, 0.1 M sodium nitrate) was destructively distilled, essentially no ruthenium was volatilized while dilute nitric acid passed overhead. However, as

the still temperature passed through the range 130 to 140°C, where the salt would be decomposing, the ruthenium concentration in the condensate rose very sharply and substantially all of it was carried away. Little additional ruthenium appeared in the vapor as the residual solids were heated to 400°C.

In further tests, the operation of a calciner was simulated by dropping a simulated waste containing tracer ruthenium onto a heated bed of alumina on a porous plate. Air or other gas was passed through the bed to simulate the calciner atmosphere. When decomposition was carried out at 400°C in an air stream, ruthenium was substantially all volatilized and carried away in the off-gas. The addition of up to 1.0 M sodium nitrite to the feed did not alter this significantly, contrary to the effect of sodium nitrite during nitric acid distillation. With a nonoxidizing atmosphere provided by a nitric oxide gas stream (estimated oxygen content about 0.014%) only about 1% of the ruthenium was volatilized. Increasing the decomposition temperature to 500°C in air reduced the ruthenium found in the off-gas to about 1/3 of that in the feed. In the latter case, much ruthenium was found to be deposited on the calciner vessel, suggesting a mechanism involving volatilization of RuO_4 followed by decomposition at high temperature. These data are summarized in Table II.

Table II

RUTHENIUM VOLATILIZATION DURING SIMULATED CALCINATION

Feed: 1.7 M aluminum nitrate, 1.0 M nitric acid, 0.1 M sodium nitrate, containing ruthenium tracer

Simulated Fluidizing Medium	Bed Temperature, °C	Calculated O_2 Content of Atmosphere, Per Cent	Per Cent of Feed Ruthenium Found	
			in Off-Gas	in Calciner and Alumina
Air	400	17	87	-
Air	550	17	29	31
NO	400	0.06	34	-
NO	400	0.014	1.4	90

Ruthenium Vapor-Liquid Equilibrium Data

Having found that ruthenium is partially volatilized under calciner conditions, it was important to determine the disposition of the ruthenium contained in the calciner off-gases upon passage of these gases through a venturi scrubber, considered for use in the removal of particulate solids from the off-gases. With regard to mass transfer of ruthenium (assumed

to be in the form of ruthenium tetroxide) from the gas to the liquid phase, the venturi scrubber should perform as essentially one theoretical stage. Under these conditions the appropriate vapor-liquid equilibrium values for the two phases define the upper limit of efficiency. It can be shown that:

$$\text{Ru Decontamination Factor} = \frac{\text{Bleed Rate}}{\text{Vapor Rate}} K + 1$$

where

$$K = \frac{\text{g Ru/g liquid}}{\text{g Ru/g vapor}} \text{ at equilibrium}$$

A few values for K were obtained at 80°C for appropriate nitric acid concentrations. A Gillespie equilibrium still[12] was modified so that a stream of air could be metered through the still pot. Appropriate measures were taken to insure isothermal conditions in the liquid system and to assay for noncondensible ruthenium. Tracer redistilled ruthenium tetroxide was used with conventional counting techniques for analysis. The variation in K with acidity is given in Fig. 2. From these data it is apparent that ruthenium transfer to the liquid phase will be high for scrub-solution and concentrations below 10 molar.

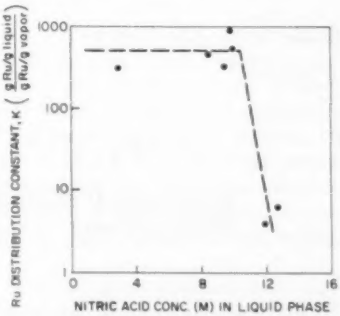

Fig. 2

Absorption of Ruthenium in Nitric Acid

Distribution of RuO_4 between nitric acid and vapor in equilibrium at 80°C. (75 mole per cent air). RuO_4 in liquid phase: 0.1 mg Ru/g liquid

Ruthenium Adsorption on Silica Gel

Laboratory studies were made to investigate the removal of ruthenium from calciner off gases by adsorption on solid surfaces. A survey of common adsorption and filtration media including commercial activated alumina, fluid bed calcined alumina, stainless steel mesh and wool and silica gel, led to the selection of silica gel as the most applicable for a dry adsorber for ruthenium. The factors most favorable for silica gel utilization were established by laboratory experiments to be as follows:

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1. Effective at vapor velocities up to 0.6 feet per second.
2. Relatively stable to the acid content of the vapor.
3. Ruthenium readily washed off with water in place at 60°C.
4. Adsorbent reactivated at 180°C in place.
5. Adsorbent readily changed and removed from the system remotely.

Decontamination factors between 200 and 1000 were indicated for beds of 24 in. or greater depth with ruthenium loading capacity of about 10 grams per cubic foot of silica gel. Operation of the bed was optimum just above the dewpoint of the macro vapor phase. Stainless steel mesh gave decontamination factors of only about 20 to 300 for a $13\frac{1}{2}$-in. bed depth and could be loaded with 0.25 gram per cubic foot of bed.

PILOT PLANT STUDIES (RADIOACTIVE)

In order to study the calcination process with radioactive liquid wastes, a shielded six-in. diameter unit was constructed at Argonne National Laboratory for semi-remote operation. The unit was designed to use diluted radioactive waste, and was capable of handling about 100 curies of 1 Mev gamma radiation. The shielding consisted of a concrete cell having 18-in. thick walls. A schematic diagram of the calcination facility is shown in Fig. 3.

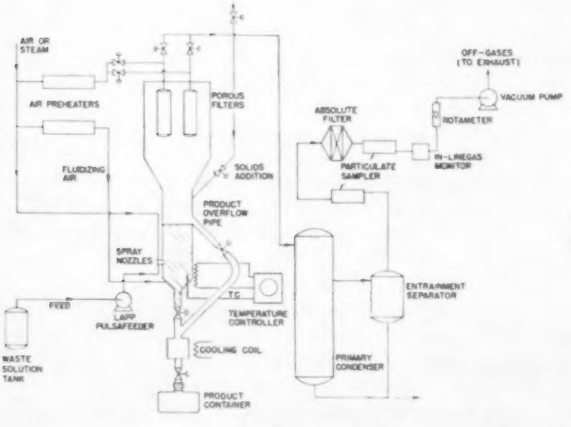

Fig. 3

Experimental Shielded Waste Calciner

The calciner was constructed entirely of type 347 stainless steel, and was similar in design to the units already described. An enlarged chamber, 12-in. in diameter at the top of the calciner housed the sintered stainless steel dust filters. The decomposition gases leaving the filters entered a

condenser, which removed most of the water vapor and a portion of the oxides of nitrogen in the form of a dilute nitric acid condensate. The noncondensible gases passed through de-entrainment traps to remove mist and then through an absolute filter (AEC type with glass-asbestos filter medium). To reduce the hazard resulting from leakage the system was normally operated under a slight vacuum supplied by either a steam jet or a vacuum pump.

Flow control valves were operated by extension handles through the cell wall. Remote operation was employed except for the occasional removal of product containers or for discharge of the bed. The activity levels permitted entrance into the cell for brief periods when necessary for maintenance or other purposes.

The operating procedure entailed charging the starting bed of inactive aluminum oxide, fluidizing, heating to the desired temperature, then introducing the solution to be processed. Cooling coils around the nozzles aided in preventing vaporization within the nozzles. With exception of bed temperature, feed composition, and fluidizing gas composition, the operating conditions were essentially constant for all runs. The nominal superficial velocity of the fluidizing gas was 1.3 ft/sec at the operating temperature and pressure. The actual gas velocity, particularly above the feed injection point, was considerably higher because of the atomizing gas and the decomposition gases. The feed rate ranged from 89 to 102 ml/min (1.4 to 1.6 gal/hr).

The feed was prepared by dilution of radioactive waste (produced at the Idaho Chemical Processing Plant and aged four years) with inactive solution of approximately the same composition. The feed contained 2.2 molar aluminum nitrate, 1.25 molar nitric acid, 0.15 molar sodium nitrate, and 0.008 molar mercuric nitrate. Initial runs were made with one part of radioactive wastes to 1000 parts of inactive solution, and the proportion of radioactive waste was gradually increased to 1 to 100 in later runs. Air was used as the fluidizing gas in most runs, but a few runs were made with other gases. Calcination temperatures in the range of 350 to 550°C were studied.

Fission product behavior was determined by analyzing the calcined solid, the condensate, and the off-gas. The off-gas was sampled for particulate activity both prior to and following the absolute filter. The off-gas was monitored just before exhaustion by passage through a one-cubic-foot box containing four GM tubes. In no case was any residual activity detected in the gas by the monitor.

Fission Product Behavior During Waste Calcination

The primary objective of the runs with radioactive waste was the determination of fission product distribution throughout the system under

various operating conditions. A secondary objective was the preparation of calcined oxide for studies on leachability of the fission products with water and dilute acids.

Results disclosed that all of the fission products in the waste were non-volatile with the exception of ruthenium. In a series of runs made at a calcination temperature of 400°C the bulk of the ruthenium activity was volatilized and carried out of the calciner in the off-gas stream. The volatilized ruthenium was efficiently removed from the off-gases along with water vapor and some of the nitrogen oxides by cooling and condensing the gas stream. The remaining trace of ruthenium activity was nearly completely removed by the absolute filter as discussed below.

The concentrations of radioactivity in the influent and effluent streams in a typical run at 400°C are shown in Table III and the distribution of ruthenium activity in a series of runs at this temperature are summarized in Table IV.

Table III

DISTRIBUTION OF FISSION PRODUCT RADIOACTIVITY DURING FLUID-BED CALCINATION OF LIQUID WASTE
(Calcination Temperature 400°C)

	Radioactivity			
	Beta	Gamma	Ru γ	Cs β
Feed Solution (cpm/ml)	6.1×10^6	1.4×10^6	1.1×10^5	6.4×10^5
Calcined Solids (cpm/g)	1.5×10^7	3.6×10^6	5.3×10^4	2.0×10^6
Condensate (cpm/ml)	1.1×10^3	5.0×10^4	4.6×10^4	1.4×10^2
Non-Condensable Gas (cpm/cu ft)	-	1.4×10^3	1.4×10^3	-

Table IV

RUTHENIUM DISTRIBUTION IN EFFLUENT STREAMS DURING WASTE CALCINATION AT 400°C

Run No.	Ruthenium Content[a] (% of input)			
	Calcined Solids	Condensate	Non-Condensable Gas	
			Before Absolute Filter	After Absolute Filter
4	-	75	0.14	0.0014
5	-	69	0.17	0.0015
7	-	75	0.13	0.0020
10	-	43	0.09	0.0013
30	20	65	-	-

[a]Some Ru deposited on interior walls of vessels and piping, causing low material balances.

Because of the refractory nature of the calcined alumina, an accurate determination of its ruthenium content was difficult. However, the results obtained indicated that about 20 ± 10% of the feed ruthenium was retained in the solid product. The ruthenium in the condensate ranged from 43 to 75% of the feed ruthenium, and the non-condensable off-gas contained less than 0.2%. The condenser was therefore effective in removing about 99.8% of the ruthenium from the calciner off-gases. The remaining ruthenium was further reduced by about a factor of 100 by means of the absolute filter.

In many runs the ruthenium which could be accounted for in the effluent streams amounted to roughly 85% of that fed to the calciner. Though some of this loss might be attributed to experimental error, much of it is believed due to plating of ruthenium in the off-gas piping.

Effect of Calcination Temperature on Ruthenium Volatilization

Since ruthenium tetroxide is known to be unstable at high temperatures, several runs were made to determine whether the volatilization of ruthenium could be reduced by increasing the calcination temperature. The results obtained at temperatures ranging from 350 to 550°C are listed in Table V.

Table V

EFFECT OF CALCINATION TEMPERATURE ON RUTHENIUM VOLATILIZATION

Temp (°C)	Ruthenium Content (% of input)	
	Condensate	Calcined Solids[a]
350	84	11
400	65	15
450	35	12
500	3	78
500	1	73
550	0.4	-

[a]Due to unavoidable losses during analyses, values reported for ruthenium in solids are known to be low.

The ruthenium volatilization decreased drastically with increasing temperature from about 90% at 350°C to less than 1% at 550°C.

In one extended run totaling 13 hours, in which the calciner was operated at three temperatures (400, 450 and 500°C), the condensate samples

showed rapid response to changes in calcination temperature. The volatilized ruthenium dropped from 65% at 400° to 35% at 450°, and to about 3% at 500°C. It is likely that at the higher temperatures the ruthenium in the feed was converted chiefly to non-volatile ruthenium dioxide, and only a very small percentage of volatile ruthenium tetroxide was formed.

Suppression of Ruthenium Volatilization with Reducing Atmospheres

It is known that ruthenium tetroxide is produced only under strong oxidizing conditions such as are present in concentrated nitrate media at elevated temperatures. An initial attempt was made to suppress ruthenium volatilization by adding 0.1 molar sodium nitrite to the feed solution as a reductant while replacing all inlet air by nitrogen. These measures were ineffective, indicating that more drastic reducing conditions were necessary to prevent ruthenium oxidation and volatilization.

Consideration was given to the use of an inexpensive gaseous reductant which could reduce all of the nitrate to nitrogen or lower-valent nitrogen oxides. A mixture of carbon monoxide in nitrogen, simulating commercial producer gas, was therefore tested in place of air as the fluidizing gas in several runs. All other air streams to the calciner were replaced by nitrogen.

The results obtained in runs made at 400°C and 550°C are compared in Table VI with runs made under the same conditions except with air as the fluidizing gas. At 400° the ruthenium volatilization was decreased from 43% when air was used to 1.0% when carbon monoxide was used. At 550° the ruthenium volatilized was only 0.3 to 0.4% with either gas. All of the nitric acid was destroyed in the runs made with carbon monoxide, and the condensate was essentially neutral, whereas in the runs made with air the condensate contained over 3 N nitric acid.

Table VI

EFFECT OF CARBON MONOXIDE GAS ON RUTHENIUM VOLATILIZATION DURING WASTE CALCINATION

Bed Temp. (°C)	Fluidizing Gas	Condensate	
		Ruthenium (% of input)	Acidity
400	air	43	3.9 N
400	CO - N_2	1.0	neutral
550	air	0.4	3.3 N
550	CO - N_2	0.3	neutral

The heat generated in the reaction of carbon monoxide with nitrates reduced the amount of external heat required for the calcination of the waste solution. However, not all of the reaction took place within the fluidized bed. While the bed was maintained at the indicated temperatures, the gas temperature above the bed rose to over 700°C. Apparently the residence time of the gas in the 20-in. deep bed was not sufficient to complete the reaction.

In addition to suppressing ruthenium volatilization, carbon monoxide appears to be useful for avoiding nitrate disposal problems and/or the corrosion problems associated with the evaporation and concentration of the nitric acid-bearing condensate. In addition the heat supplied by the reaction would decrease the external heat requirement. The feasibility of using carbon monoxide must await experiments in larger-scale equipment, which will provide additional bed depth and additional residence time for the gas.

Performance of the Sintered Metal Filters

The sintered stainless steel filters, which had a mean pore diameter of 10 microns, were found to be highly efficient for the removal of dust from the calciner off-gases. These filters, when coated with a dust layer, were capable of removing sub-micron sized particles. Filter efficiency was evaluated by determining the amount of non-volatile (non-ruthenium) activity in the condensate and in the non-condensable gases. Non-volatile radioactivity could be present in these streams only by passage through the filters in the form of solid particulate.

The results obtained in several runs are presented in Table VII.

Table VII

NON-VOLATILE RADIOACTIVITY CARRIED THROUGH SINTERED METAL FILTERS IN WASTE CALCINER

Run No.	Non-Volatile Radioactivity (% of feed)	
	Condensate	Non-Condensable Off-Gases
33	2.5×10^{-3}	-
40	2.2×10^{-3}	5×10^{-5}
41	1.6×10^{-3}	8×10^{-4}
42	2.2×10^{-3}	3×10^{-5}

These data represent the average values for several samples taken during the course of the runs. The non-ruthenium activity which passed through the sintered metal filters was about 2×10^{-3}% of the activity fed to the calciner. Most of this radioactivity was removed in the condenser and only

about 1×10^{-4}% remained in the non-condensable gas. Further removal undoubtedly occurred in the absolute filter, but the amount could not be determined because the activity level was too low. The radioactive nuclides present in the condensate and gas samples after separation of ruthenium were identified as cesium-137 and cerium-144, which are the predominant gamma emitters expected to be present in the aged wastes.

Experience with the sintered stainless steel filters in the experimental calciner indicates that good performance could be expected from these units in a plant-scale installation. Corrosion was encountered only in the runs made with carbon monoxide gas, during which holes or cracks developed in several of the bayonet filters. In these runs the gas temperature was so high (770°C) that reaction of the carbon monoxide with the steel might be expected. Long term corrosion tests would be required before the filters could be considered suitable for plant installation. Cooling the off-gases before filtration, possibly by spraying water into the space above the bed, would be desirable to minimize corrosion.

To demonstrate that the filters can be operated over a long period without plugging, a test facility was installed, consisting of a 6-in. diameter glass pipe containing a bed of fluidized 20 to 325 mesh alumina. Two sintered metal filters were installed above the bed for filtering the gas. These filters were automatically cleaned on a pre-set time cycle by a high-velocity, short-duration, jet of air directed into the filter discharge opening through a venturi-shaped fitting. Solenoid valves operated by a timer provided a 1/2-sec pulse of air once each minute for each filter from a line pressure of 100 psig. The pressure drop across the filters showed an initial increase of about 6 in. of water during the first few days of operation, then remained essentially constant at 40 inches of water over a period of 30 days of continuous operation. This test proved that plugging could be avoided by this method of cleaning, which permits a pre-coat of alumina dust to cover the filters at all times and removes only the outer layer of dust during blowback.

Concentration of the Condensate

Data already presented has shown that partial condensation of the calciner off-gases is an effective method for removal of both the volatilized ruthenium and the particulate radioactivity from these gases. This method produces a low-level, dilute-nitric acid waste requiring further treatment before disposal. Cursory experiments were carried out to determine whether this solution could be concentrated by evaporation without excessive ruthenium carryover. It was found that direct evaporation to high nitric acid concentrations (above 10 M) resulted in appreciable volatilization of ruthenium. However, the use of a continuous equilibrium evaporation method, in which the feed was diluted with water to a concentration of 1.5 M nitric acid,

allowed the still pot acid concentration to be maintained at 7 to 8 M. Under these conditions very little volatilization of ruthenium occurred.

GENERAL PROCESS CONSIDERATIONS

Equipment Reliability

For plant use, it is imperative that this type of equipment operate reliably for long periods without maintenance. The 6-in. pilot-plant calciner has operated for over 3000 hours in runs of varying duration. The longest continuous run extended for 548 hours and was terminated voluntarily. Extended operations for up to several days are normally required for the bed to reach a nominal equilibrium condition. Many runs of this nature have been made followed by rapid turn-around and start-up under modified conditions. Rarely has trouble been experienced with the formation of large aggregates in 6-in. and larger calciners. Large particles have been formed irregularly in smaller diameter units.

Materials of Construction

Material requirements for the calciner are in general not very severe. Ordinary steel appears to withstand the corrosion and erosion conditions in the body of the calciner as long as the atmosphere is hot and dry. However, since decontamination of a piece of radioactive equipment is normally required at intervals, stainless steel, Carpenter 20, and carbon steel were compared in tests run in a 3-in. diameter corrosion test calciner for over a thousand hours. Weight losses were only of a few mils per year or less; however, scaling was observed on the carbon steel surface. Some corrosion was observed in stainless steel feed nozzles in early tests; titanium alloy inserts have since performed very well in this service. Considerable erosion was observed in stainless steel jets used to return fines from the scalp cyclone. Very hard inserts of boron carbide and similar material markedly reduced this problem.

Heat Transfer and Capacity

The rate of heat transfer between a water-cooled test loop and the fluidized bed in the 6-in. calciner has ranged from 25 to 250 $Btu/(hr)(ft^2)(°F)$, with typical values being about 80. The higher rates were obtained when the bed consisted primarily of small particles. When large particles were produced electrical heater failure occurred due to inefficient heat transfer, causing element burnout. Data have not yet been obtained for bed heating by combustion gases or liquid metal. The capacity of the systems are limited primarily by the ability to introduce heat to the bed, and by the off gas volumes which can be handled following the calciner. The 6-in. calciner has

been operated at rates from 20 to 220 ml/min. The 4-sq ft calciner has been operated at a rate of 30 liters per hour using a direct-fired tube heater with 1/3 the transfer surface of the liquid metal heater (the latter was designed for 100 liter/hour).

Characterization of Alumina Produced by Calcination

Alumina produced by fluidized bed decomposition of aluminum nitrate consists of white, nearly spherical particles. Under high magnification, porosity and a multilayered structure can be observed.

An x-ray diffraction study of a sample of alumina produced at 400°C indicated that two-thirds of the material was alpha-alumina and the remainder was amorphous. Examination of samples produced in a laboratory-sized calciner (3-in. diameter) indicated that the alpha phase constituted only 30% of the bulk particles and as little as 10% of the fines. Ignition of samples of aluminum nitrate, dibasic aluminum nitrate, and aluminum hydroxide in a muffle furnace at 400°C produced completely amorphous particles. Partial conversion of both nitrate compounds to alpha alumina occurred between 400 and 800°C, and all compounds were substantially converted to alpha-alumina between 800 and 1100°C. This suggests that the dibasic nitrate rather than the hydroxide may be an intermediate in the calcination process. The form of alumina present will probably affect the resistance of particles to attrition and the ability to remove soluble components by leaching.

ASPECTS INVOLVED IN STORING THE CALCINED WASTE

Leaching Fission Products From Calcined Wastes

Studies of the dilute nitric acid leaching of long-lived heat-producing radioactive isotopes from calcined alumina, obtained by decomposing aluminum nitrate containing about 1% of full activity waste, indicate that practically all the cesium, perhaps 3/4 of the strontium, and 10% of the cerium can readily be removed in multiple contact operation. This would have little effect on the heat generation rate of calcined solids in the first few years of decay, but with wastes aged 5 years the leached solids might generate heat at only about 1/3 the rate of unleached solids, as shown in Fig. 4. Ruthenium and zirconium-niobium are only slightly extracted. Any soluble salts present, such as sodium nitrate, are extracted at the same time along with a few per cent of the alumina.

Perhaps more significant is the reduction in the availability of biologically-hazardous fission products to contact with ground water which may be effected by leaching before storage. For example, in a 5-stage batch leaching the last solution equilibrated with calcined waste had a strontium-90

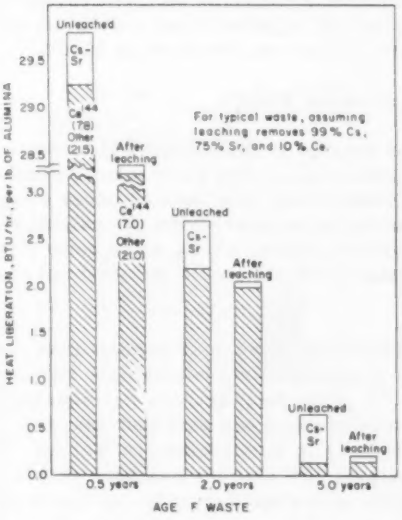

Fig. 4. Effect of Leaching on Decay Heat in Solid

content which was only 1/500th that of the initial leach solution. The extraction of cerium appears to be more time-dependent and increases with long contact.

Leach solutions, or concentrates prepared for them, would require storage and containment separately, but in a volume only a few per cent of the initial waste. Such solutions could be used for separation and isolation of fission products for source preparation. Since strontium is a major component, it is probable that preparation of a separate package for long safe storage of this would be desirable.

Thermal Conductivity of Calcined Alumina

Estimation of the rate of dissipation of heat from calcined alumina containing decay fission products in storage requires a knowledge of the effective thermal conductivity of the solid material at elevated temperatures. It was anticipated that the conductivity of alumina produced by waste calcination would differ from that of alumina for which data are available due to the way in which the particle is built up and to the effect of other waste components, such as sodium compounds, which may remain with the alumina. At higher temperatures, transfer of heat by radiation would also occur significantly. Measurements were made inserting thermocouples at intervals in centrally heated spherical and cylindrical beds of the calcined product. The apparent conductivity increases almost linearly from 0.08 to 0.23 Btu/(hr)$(ft^2)(°F/ft)$ between 100 and 1000°F and further increases to 0.30 Btu/(hr)$(ft^2)(°F/ft)$ at 1740°F.

Fission Product Volatilization from Calcined Waste at High Temperatures

The central temperatures in a stored mass of calcined waste could reach very high values, depending upon the method of storage and the rate of heat dissipation. To determine at what temperatures the volatilization of fission products might occur, samples of the oxide produced in the pilot-plant calciner were ignited in a muffle furnace at temperatures between 700 and 1200°C. No loss in radioactivity was observed in two-hour tests at temperatures up to 1000°C. At 1200°C about 85% of the gamma activity was

volatilized, probably by decomposition of cesium oxide to volatile cesium metal. Examination of the ignited samples with a gamma-ray spectrometer showed that virtually 100% of the cesium activity was removed at 1200°C.

Permanent Fixation of Radioactivity in Alumina Waste

Although initial plans call for the storage of the calcined alumina in metal vessels, an alternative approach involving a more permanent method of fixing the radioactivity is also being considered. One method under study at the Massachusetts Institute of Technology is to incorporate the calcined solids into a high-alumina ceramic glaze formulation which, after being fired to a vitrified mass, would retain the fission products with a minimum of additional protection.

The effect of standard ceramic parameters, such as composition ratios, on glaze firing temperatures and fission product leachability have been studied. A leaching rate of 8×10^{-4}% of the total activity per square centimeter per week was obtained with a mixture of one part alumina, one part boria, one part calcia, and 3½ parts silica in small spherical pellets about 1/2 in. in diameter. A glaze containing 23% alumina has been developed which melts at 1300°C. Volatilization of cesium during firing has been encountered in many experiments.

Attempts have also been made at Argonne National Laboratory to apply ceramic glaze coatings to the individual particles of calcined waste by spraying the ceramic in slurry form into a fluid-bed of the alumina in a manner similar to the calcination process itself. Successful coatings of several mils thickness have been obtained; but poor glazing resulted after firing, probably because of reaction of the alumina with the thin ceramic layer.

DEMONSTRATION PLANT

Preliminary engineering designs of a 60 gal/hr demonstrational facility is nearing completion. This facility is designed to develop engineering information and will be capable of calcining wastes generated from processing fuels at 200 days post reactor discharge. Direct maintenance of the process equipment has been a major design criteria. Therefore, the primary consideration has been to utilize mechanically simple equipment which will require a minimum of maintenance and can be easily operated behind the required shielding. The simplified flowsheet for the process is shown in Fig. 5. The basic components of the process are (1) the calciner and its auxiliaries, (2) off-gas treating equipment, and (3) solids storage facilities.

The fluidized bed calciner is designed to operate under a slight negative pressure at the design rate and a temperature of 400°C. The calciner

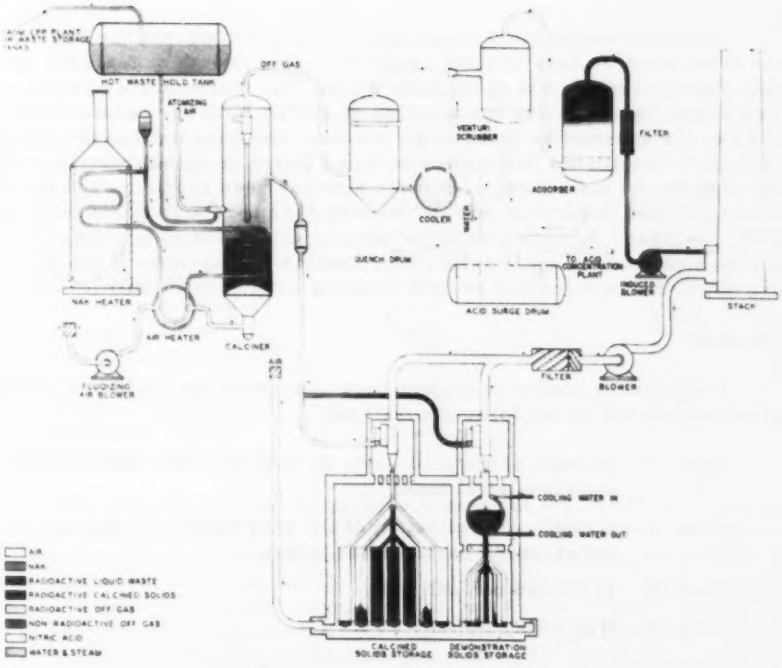

Fig. 5. ICPP Radioactive Waste Calcination Plant

will be sufficiently flexible to permit operation at 500°C with approximately 2/3 of the design feed rate. Calcined alumina will be pneumatically transported to the solids storage bins by an air stream which will be returned as part of the fluidizing air. Local air tolerance levels are such that the nitrogen oxides can be discharged to the atmosphere provided they are within the tolerance limits on radioactivity. It is recognized that a simple but highly efficient off-gas system will be required to avoid emission of radioactive particulates to the environment. Therefore, the system chosen for the preliminary design essentially consists of several air cleaning devices in series. Upon leaving the calciner the off-gases will pass through high efficiency cyclones, wet scrubbers, adsorber-filters packed with silica gel, and absolute AEC-type filters before joining the building ventilation air for stack discharge to the atmosphere. The system chosen is mechanically simple and involves a minimum amount of remote removal equipment. For example,

the silica gel beds will be pneumatically removed after they have exceeded their regenerable life, and will be incorporated with the calcined solids for storage. The final filters will be the only remotely removable and replacable piece of process equipment.

Sufficient solids storage volume will be provided with the facility to store three hundred days of plant production. The design criteria for the solids storage system was to limit the center line temperature to a value not exceeding the calcining temperature of 400°C. This temperature was chosen on the assumption that fission products not volatilized in the calciner would not be volatilized in the storage bins. Based on these criteria a design employing forced air cooling of storage bins has been adopted. The solids will be contained in narrow vertical annular chambers with intervening air cooling passages. A large number of temperature measuring points will be installed since the design is sufficiently flexible to determine if temperatures above 400°C can be tolerated without creating an off-gas problem.

Economics

Preliminary economic studies have been made for disposing of aluminum nitrate wastes by the following methods.

Case I. Storage of the acid waste in water-cooled, underground stainless steel tanks.

Case II. Caustic neutralization of the acid waste with storage in underground carbon steel tanks.

Case III. Fluidized bed calcining.

Case IV. Ball mill calcining.

The estimated investments and operating costs have been developed from known and estimated costs of construction at the National Reactor Testing Station. The topographical conditions and requirements of this site are such as to make construction relatively expensive, therefore, it is believed that these estimates probably represent near maximum costs.

In preparing the cost estimates no long-term storage costs, such as cooling water circulation, surveillance, etc., have been included since it is believed that in all four cases these costs will be approximately the same on the basis of an initial gallon of liquid waste. Recent cost estimates for Cases I and II indicate that total initial investment plus chemical treatment will cost, repectively, $2.40 and $2.62 per gallon of original acid waste.

For Cases III and IV the following table has been developed on the basis of a plant capable of processing 200 gal/hr of 200-day post-reactor-discharge waste for three hundred days per calendar year. The plant

investments are exclusive of the solids storage system but include, in addition to the calcination equipment, various process components for recovery or cleaning of off-gases and solids leaching prior to commitment to final storage.

	Case III	Case IV
Plant Capacity	1,440,000 gallons per operating yr.	1,440,000 gallons per operating yr.
Estimated Plant Investment (excluding solids storage)	$7,025,000	$10,300,000
Direct Operating Costs	$/gallon	$/gallon
Labor	0.101	0.121
Utilities	0.111	0.212
Total Direct Operating Costs	0.212	0.333
Indirect Operating Costs	0.045	0.054
Total Operating Costs (excluding depreciation and solids storage)	0.257	0.387
Depreciation (15 years)	0.323	0.476
Total Processing Costs (excluding solids storage)	0.582	0.863

Preliminary cost estimates have been made for solids storage systems capable of containing the calcined solids in cooled rectangular bins of such dimensions as to prohibit centerline temperatures exceeding 400°C for solids generating 23.8 Btu/(hr)(lb) (see Fig. 4). The bin thickness is estimated as approximately nine inches and the investment is estimated as $1.238 per gallon of original waste. For the same centerline temperature a 51-in. thick bin is estimated for solids generating 0.7 Btu/(hr)(lb) and the investment is estimated as $0.39 per gallon of original waste. These estimates have been based upon a waste which when calcined would generate approximately 0.75 pound of solids per gallon of original liquid. Latest research and development data indicate the probability of successfully storing solids at much greater centerline temperatures with concomitant reduction in storage costs. Also the short-term liquid storage of green waste to permit removal of the initial high heat evolution may prove economic.

Others[13] have discussed in great detail the effect of waste disposal costs upon the economics of nuclear power. Many of the advantages of solids storage of radioactive wastes such as the safety aspects are very difficult to evaluate economically. However, it is apparent that the fluidized bed calcining of radioactive wastes should be economically competitive with liquid storage and less hazardous.

Application of Process to Other than Aluminum Nitrate Wastes

The fluid bed conversion process is felt to be generally applicable to the production of solid wastes from solutions by the removal of water and the decomposition of salts. In order to produce a stable solid in which fission products will be effectively retained, it appears desirable that the wastes yield primarily insoluble oxides such as those of most di- and polyvalent metals. Nitrate salts are most easily decomposed but salts of other acids can probably be processed with appropriate modifications.

If oxide-forming compounds or other high temperature-stable compounds are not initially present in the waste, such compounds might be added to the solution before calcination. Clay, silica, or other ceramic materials might form suitable solid media for the deposition of stable solids from Purex-type wastes containing little or no metallic oxides other than the fission products.

Some preliminary tests have been made on the conversion to solid form of wastes simulating those produced in processing enriched zirconium and stainless steel fuel compositions.[14] These tests were carried out in the 6-in. pilot plant calciner at the NRTS. These fluoride and sulfate-containing wastes were readily converted to granular solids. However, the solids produced contained a considerable proportion of water-soluble materials. Many successful runs have also been made at ANL on the conversion of zirconium fluoride solutions to dry, granular zirconyl fluoride.

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Representative HOLIFIELD. Our next witness is Mr. W. B. Heroy, of the Geotechnical Corp., Dallas, Tex.

You may proceed, Mr. Heroy.

STATEMENT OF WILLIAM B. HEROY,[4] THE GEOTECHNICAL CORP., DALLAS, TEX.

Mr. HEROY. I am here in two capacities. One is as a consultant to Oak Ridge National Laboratory, and the other as a member of the Committee on Waste Disposal of the National Research Council. I will primarily speak as a consultant to Oak Ridge.

1. This paper considers the possibilities of the disposal of high level waste in underground cavities produced by excavation or solution and the program of research and development by which these possibilities are being evaluated.

High level waste results from the chemical processing of the spent fuel elements removed from a nuclear reactor. The raw waste has three important characteristics:

1. It is highly radioactive;
2. It produces heat;
3. It is strongly acid.

Various methods for the solidification of the waste for easier storage have been developed experimentally. You have heard several of those, of course, described this morning. All methods of solidification introduce into the disposal problem an additional element of cost which would not be required if the waste can be safely and economically transported and stored in the liquid state. For these reasons, the experimental program described in this paper is predicated on the storage of waste in liquid form.

Two principal methods of underground containment in geological formations are considered:

1. Storage in sedimentary rocks by means of deep wells; and
2. Storage in underground openings, either natural or artificial, at comparatively shallow depths.

The problems encountered in the disposal of radioactive waste in deep wells have been examined by a committee of the American Petroleum Institute, and will be discussed in a paper to be presented here by its chairman, Dr. Moore.

There are several alternative possibilities for storage beneath the surface. Storage in quarries is considered to be unreliable because such openings are rarely watertight and because some means of sealing would be required to protect shallow underground waters. In natural caverns and shallow mines the problems of tightness and leakage are about the same as in surface excavations. Another possibility is storage in deep mines, below the fresh water aquifers. Various types of deeper mines were considered. Coal and metal

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Work history: Junior and assistant geologist, U.S. Geological Survey, 1907–12; geologist, Land Classification Board, 1913–18; chief, Division of Power Resources, 1918–19; advisory engineer, U.S. Fuel Administration, 1918–19; geologist, foreign manager, Sinclair Oil Corp., 1919–28; chief geologist, manager, exploration department, 1929–38; president, Pilgrim Exploration Co., 1939–42; Director, Division of Reserves, Petroleum Administration for War, 1942–43; Director, Division of Foreign Production, 1943–46; Special Assistant, Secretary of the Interior, 1946; vice president, the Geotechnical Corp., 1946–52; president, 1952–; member, Advisory Committee, U.S. Geological Survey, 1946–; member, Earth Science Panel, National Science Foundation, 1956–.

mines are usually wet because of the entrance of water along joints, veins, and fissures. Coal mines usually contain explosive gases that require removal by constant ventilation. Under these conditions, the containment of liquid radioactive waste would be a difficult problem. Space in salt domes or salt beds, or in abandoned salt mines, is considered to be the most promising environment. Special studies of the possibility of storage of waste in salt space have been carried out under the direction of Oak Ridge National Laboratory. The research has covered all of the problems related to waste disposal in salt that could be foreseen. Dr. E. G. Struxness will present a paper which describes the investigations that have been made with regard to the relation of radioactive waste to a salt environment.

2. Characteristics of rock salt: I will summarize engineering and geological factors that characterize salt and salt space.

Rock salt weighs about 135 pounds per cubic foot. That is approximately 15 cubic feet per ton. Its melting point is high, 1,475° F., and it has a much larger thermal conductivity than other common rocks. It is nonporous and impervious to the flow of liquids and gases, although some limited movement of fluids may occur along the faces of the salt crystals. It is highly soluble in water, and is chemically very stable. I assume that possibly some in the audience may not have seen a piece of rock salt. The upper surface of this core shows the crystals of the salt in their condition as they are mined. This will give you an idea of how solid and heavy a rock it is.

Salt has considerable compressive strength, but is also plastic. At normal temperatures its movement is controlled by plastic flow, and deformation occurs slowly when salt is under pressure below the yield point. The pressure at which salt will yield and fracture varies somewhat, but tests indicate that this occurs within a range of 2,500 to 4,000 p.s.i. By "slowly" I mean very slowly, a matter of years. You can see from the sample that it is a firm and solid rock.

These unique characteristics of salt give deposits of the rock remarkable geological features. Because of its imperviousness, excavations in salt are practically always dry, even though the salt is overlain by water-bearing strata. Because of its plasticity, any fractures in salt close rapidly and jointing is absent. Because of the strength of the rock, large spaces may be mined out, provided that pillars of sufficient size are left so that the weight of the overlying rocks do not exceed the breaking strength of the salt. Even at depths of a thousand feet, two-thirds of the salt area may be removed without perceptible deformation of the pillars.

In bedded salt, partings of shale and anhydrite are usually present. Beds 10 feet in thickness without such partings occur over large areas, and beds up to 30 feet in thickness without partings are presently being mined. In salt domes, those of the type that occur on the gulf coast, the salt may cover areas of several square miles and extend to great depths. The salt itself is generally quite pure, and most of the rock salt mined contains less than 3 percent of impurities. This is important, of course, because of the possible chemical impurity reaction with the waste.

3. Distribution of rock salt in the continental United States: As shown on the accompanying map, figure 1, deposits of rock salt underlie large areas within the United States, the total extent being of the order of 400,000 square miles, one-seventh of the total land area, ex-

cluding Alaska. Salt occurs at economically workable depths within three principal areas. The Silurian area is situated in the northeastern part. The salt occurs in two basins connected by a narrow neck. The Salina, or Appalachian basin includes parts of New York, Pennsylvania, West Virginia, and Ohio. The Michigan basin includes nearly all of Michigan and part of the province of Ontario, Canada. The Permian salt basin lies in Kansas, Oklahoma, Texas, and New Mexico. The gulf coast basin extends from Alabama westward, and includes parts of Mississippi and Arkansas, and most of Louisiana and the eastern part of Texas. It extends southward under the Continental Shelf. Its salt is of Jurassic age or older. A large basin with salt of Devonian age has been found by deep drilling in North Dakota and Montana, and extends northward into Canada. In all these areas most of the salt is at depths that are too great for economical mining, but it may be produced by solution by means of deep brine wells.

In the Silurian and the Permian basins the salt occurs in bedded deposits that are essentially flat lying. Rock salt is mined in New York, Michigan, and Kansas. In the gulf coast, rock salt is found in thick bedded deposits at great depths. Under the pressure of the overburden, same of the salt has been forced upward through the overlying beds as great intrusive masses which are called salt domes. Over 200 of these domes are known. Many of the domes are deep seated, but others extend to the surface. Rock salt is mined from six salt domes in Louisiana and Texas.

In the basins where the bedded deposits have been developed or explored by deep wells, salt has been found to be present in numerous beds, and with great thickness. In New York, some deep borings have penetrated over 500 feet of salt in several beds. In Ohio, the maximum thickness is about 300 feet, with individual beds 50 feet thick. In the center of the Michigan Basin, the total thickness of salt reaches 1,800 feet. The salt in the Kansas part of the Permian Basin reaches a maximum thickness of about 400 feet, and over 200 feet is present in several beds over large areas. In the West Texas-New Mexico part of the Permian Basin, the aggregate thickness of the salt is over 2,000 feet at the maximum, and is over 500 feet under thousands of square miles.

An immense area in the gulf coast geosyncline is probably underlain by bedded salt at great depths. Few wells have penetrated the salt formation. The salt thickness drilled may not represent the thickness originally deposited because of flowage of the salt. The original thickness over large areas was probably 500 to 1,000 feet, and may have been even greater. Where salt domes have been intruded into the overlying strata, it is known from geophysical surveys and from drilling that in many cases, domes have risen over 10,000 feet above the source bed. Most of them are several square miles in area. Salt domes are also present over a large area of the submerged Continental Shelf.

In the West, drilling in recent years has disclosed the presence in North Dakota and Montana of a large deposit of bedded salt in the Williston Basin. The salt is found in strata of several ages, the oldest and thickest bed being of Devonian age. This bed has a maximum thickness of over 400 feet, and is found at a depth of 10,000 feet or more.

The Paradox Basin in western Colorado and eastern Utah is an area of anticlinal folding in which salt from a thick, deep seated salt formation has been extruded upward along the axes of the folds, forming salt structures similar to salt domes. In a few places the salt has

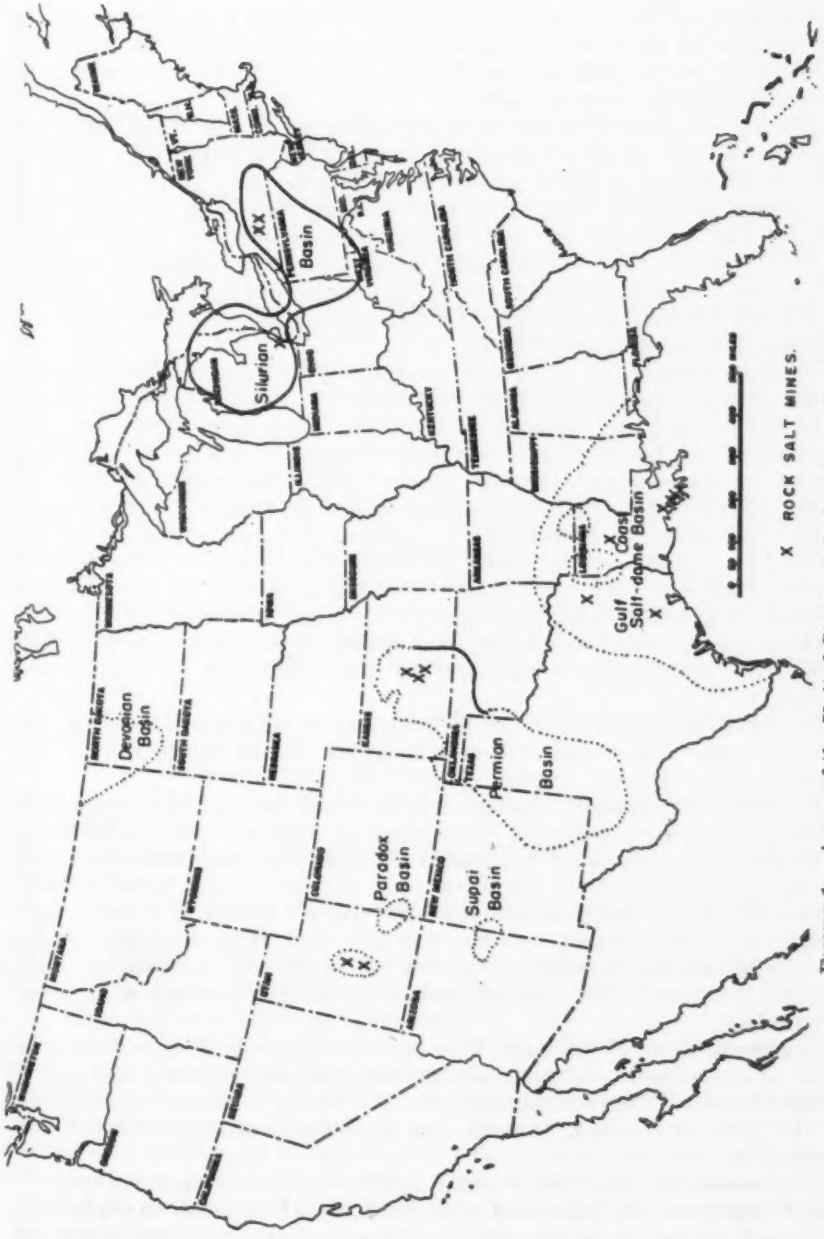

FIGURE 1.—Areas of the United States underlain by rock-salt deposits.

come within a few hundred feet of the surface, but it is generally found in wells at depths of over 5,000 feet.

It should be evident from the foregoing summary that the rock salt resources of the continental United States are of immense volume and extent. While no one has attempted to make a detailed estimate the volume of rock salt in these various basins cannot be less than 20,000 cubic miles, and may be twice that amount. In other words, it is one of our most common rocks.

The entire rock salt production of the United States, 5,341,586 short tons in 1957, is nearly all obtained from 12 large mines. In volume, this amount is equivalent to about 1,800 acre-feet. I presume you gentlemen from the West know that an acre-foot is an acre one foot deep. The quantity of salt annually produced as brine—1957, 14,518,744 short tons—is, roughly, three times the volume produced as rock salt, so that our total annual production of salt is of the order of a small fraction of a cubic mile. In comparison with our total resources, this amount is negligible.

4. Space requirements for waste storage: The volume of radioactive waste that will be produced in the future will be established by many variable conditions, some of which have been determined by past experience, while others can only be estimated or projected. Making liberal estimates as to what will be our total power required over the next 40 years, as to the total amount of energy that will be produced from nuclear fuels, and as to the amount and concentration of the waste, the volume of high activity waste that would be produced in the year 2000 is computed to be about 160 acre-feet, or less than 10 percent of the salt space now being mined out annually.

Over the 40-year period the total waste production might be five times this amount, or about half of the salt space now produced annually.

In other words, if we were to continue to produce waste for the next 40 years we could still put it away in the space mined out now in a year in salt mines.

It, therefore, appears that if only a small part of the space now being produced annually by the mining of rock salt were to be made available for storage of radioactive waste, the requirements could be met at a low cost, as the mining of the salt would have already borne the cost of the production of the space. It would even be possible to mine out entirely new space in salt, especially designed for the storage of radioactive waste, at a very moderate cost, probably at only a small fraction of the present cost of providing storage at or near the surface.

Representative HOLIFIELD. You are considering this not on the basis of the present volume, but on the basis of reducing it to size?

Mr. HEROY. That is right. It could, however, be stored on the basis of the present volume, that is, the high level waste, within these figures.

Of course, the low level waste would be a much larger volume.

5. Considerations affecting the location of a field experiment: Although extensive laboratory experiments have been conducted in the behavior of salt in relation to nuclear waste, it is realized that all of the principal factors cannot be conclusively determined in the laboratories, but must be established by means of field experiments. Such an experiment in salt storage should be conducted under con-

ditions that would resemble as closely as possible those that would be present in an actual storage operation.

In considering the location of such an experiment, preliminary thought was given to the broader problem of where radioactive waste would probably be produced in the future, and to the requirements for the transportation of waste from the points of production to the facilities for storage. It appears that most of the spent fuel from power reactors will be produced in the northeast quadrant of the United States—excluding Alaska—and that most of the waste produced from plutonium production will be in the northwest quadrant. It has been suggested that additional large scale processing plants for spent fuel will be required in the future, and that the first plant should be located in the north central area, which is, of course, the center of gravity of nuclear power production as it is now set up.

The transportation, either of spent fuel or of radioactive waste, is a hazardous procedure and is also costly because of the expense of shielding and guarding the radioactive materials. Economic studies indicate that if the reprocessing plant could be located at or near the location of the waste disposal facilities, thus minimizing the need for transportation of liquid waste, the optimum transportation situation could be realized.

In other words, it would be possible to have the reprocessing plant and the storage plant essentially in the same place to save the transportation of the highly radioactive waste.

6. Survey of possible experimental sites: In selecting the most desirable location for a field experiment in waste storage, other factors have necessarily been considered. Some of these are:

(*a*) The willingness of the management to house the experiment;

(*b*) The facilities that would be available for construction and operation of the test;

(*c*) The accessibility of the location;

(*d*) The costs that would be incurred under the local conditions.

Technical considerations that affect such a determination include:

(*e*) The depth of the salt and the corresponding weight of overburden;

(*f*) The thickness and character of the salt bed mined;

(*g*) The nature of the roof and its permanency;

(*h*) The stability of the pillars;

(*i*) The character of the rock section below the mine floor;

(*j*) The stability of the mine floor;

(*k*) The character of the salt section in which the final experiment would be emplaced.

With these factors in mind, detailed consideration has been given to all of the possible locations in the northeastern quadrant of the United States, in most cases with the benefit of field examinations of the sites themselves.

On the basis of the field surveys, it was determined that mines at Detroit, Mich., and Hutchinson, Kans. are preferable to other locations as regards physical conditions.

A full evaluation of these two possible sites was made. The conditions at Hutchinson are considered to be somewhat more favorable

than those at Detroit, and negotiations are in progress with the management of the Hutchinson mine to reach an agreement on the basis for conducting a storage experiment using simulated non-radioactive waste.

7. Possibilities of storage of solid waste in salt space: At an earlier point in this discussion it was stated that, other factors being equal, it would be economically advantageous to store the radioactive waste in liquid form, as the solidification of the waste would be an added cost. If, however, the waste could be economically solidified, the cost of excavating tanks within the salt would be obviated.

The storage of waste in solid form in salt space may have other possible advantages:

(*a*) The radioactive materials stored would be more readily available for recovery and use in case some isotopes in the waste should have future value, not now foreseen;

(*b*) The solid material should be more readily removed from storage if, in the distant future, hazards should develop that are not expected.

The storage of waste in solid form may be under the disadvantage that its transportation by mechanical means from the surface to the underground location may be more hazardous to persons than the transportation of liquid waste, which could be moved through pipes.

With regard to the cost of excavation of waste tanks in salt space, it is expected that, in an actual storage operation, the cost should be low. In commercial salt mining, the average sale value of the product is about $6 per ton. This includes processing, marketing, and administrative costs, so that the actual cost of mining is much smaller. It seems probable that, for a large-scale storage operation, the cost of providing space in underground tanks would be less than 20 cents per cubic foot. That represents, of course, only 3 or 4 cents a gallon. It is, therefore, likely that the cost of storing liquid waste in underground tanks in salt would be less than the combined cost of solidification of the waste and of mechanical movement to storage areas.

8. Possibilities of storage of liquid waste in solution cavities in salt: In the deeper portions of the geological basins drilling has demonstrated that thick beds of salt are present over large areas. As salt is readily soluble, it is being produced in the form of brine at numerous plants in the continental United States. The salt produced in that way from brine, sells for about $4 a ton with all these other costs on it, so that mining by brineing is very cheap. Wells are drilled into the salt, and water is circulated until cavities are formed. The resulting brine is removed by pumping, and this process may be continued for many years, resulting in the formation of large caverns. These are kept filled with brine, and the overlying rocks are thus supported against caving.

Another application of this process is the formation of underground storage tanks in which petroleum liquids are stored. The underground cavities are formed by solution, and the brine is then withdrawn and gasoline or other products are pumped in. As the petroleum products do not dissolve the salt, this method produces a cheap and safe storage for these flammable products. Many such storage plants are now used by the petroleum industry.

. The use of this method has been proposed for the underground storage of radioactive waste. The experimental work to be undertaken in underground tanks in salt should give information that will help to

determine whether or not the temperature of the waste can be kept below the point of boiling in solution cavities. If the radioactive waste were to boil in an underground cavity and pressure of radioactive vapor built up, a hazard might be created.

It is also difficult by the solution method to manage the shape and extent of the solution cavities and to insure that, in larger cavities, the roof will be supported against collapse. The falling of the roof of a canyon containing radioactive storage might have serious consequences. The difficulties of monitoring an experiment in the storage of waste under these conditions are quite formidable.

If, as a result of further experimental work, these problems can be resolved, the development of solution cavities in salt in the deeper portion of geological basins may have advantages over storage in deep porous reservoirs.

On the basis of this investigation we are now proceeding with an attempt to find out some of these things we want to know about the behavior of waste in salt.

Representative HOLIFIELD. Your presentation is very interesting.

Mr. HEROY. Thank you, sir.

Representative HOLIFIELD. We will ask Mr. Moore, the next witness, to appear as the first witness at 2 o'clock.

Unless there are questions of Mr. Heroy we will adjourn at this time until 2 o'clock.

(Thereupon, at 11:55 a.m., the subcommittee was recessed, to reconvene at 2 p. m. same day.)

AFTERNOON SESSION

Senator ANDERSON (presiding). The meeting will be in order. I hope Mr. Holifield will be here, but we will start in his absence.

The first witness is Mr. T. V. Moore of the Standard Oil Co. of New Jersey.

Mr. Moore.

STATEMENT OF T. V. MOORE,[5] STANDARD OIL CO. OF NEW JERSEY

Senator ANDERSON. Will you state your name for the record?

Mr. MOORE. My name is T. V. Moore. I am in the Producing Coordination Department of the Standard Oil Co. of New Jersey.

The petroleum industry, as a result of its work to improve production practices in the interest of conservation, has developed an extensive technology of injecting fluids into deep wells. Because this

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Virginia Military Institute, B.S. 1925.
Rice Institute, BA, 1926, Ch. E. 1927.
Massachusetts Institute of Technology, M.S. in Ch. E. 1920.
Work history:
Humble Oil & Refining Co.:
Development department, Baytown, Tex., 1927–28.
Leave of absence, 1928–29.
Production research division, Houston, Tex., 1929–33.
Head of production research division, Houston, 1933–42.
Loaned on special assignment, 1942–45.
Research Laboratory of Applied Chemistry, M.I.T., Cambridge, Mass., 1928–29.
Metallurgical Laboratory, University of Chicago, Chicago, Ill., 1942.
Sec. 11.3 N.D.R.C., Massachusetts Institute of Technology, Cambridge, Mass., 1943–45.
Esso Research & Engineering Co. (formerly Standard Oil Development Co.), New York, N.Y., coordinator of production research, 1945–56.
Standard Oil Co. (New Jersey), New York, N.Y., producing coordination department, 1956 to date.

technology might have some application to the disposal of radioactive waste, the division of production of the American Petroleum Institute was requested to appoint a small committee to review the problems involved; to consider, in a preliminary way, the feasibility of the method; and to call attention to those practices of the petroleum industry which may be useful in such a disposal system.

I was appointed chairman of this committee, the other members being—

C. I. Alexander, Magnolia Petroleum Co.
Gerhard Herzog, the Texas Co.
W. S. Morris, East Texas Salt Water Disposal Co.
J. W. Watkins, U.S. Bureau of Mines.
Milton Williams, Humble Oil & Refining Co.

This group met on several occasions with members of the staff of the Atomic Energy Commission, the Hanford Atomic Works, and the Oak Ridge National Laboratory to discuss the problems of radioactive waste and the present practices for its disposal.

In October 1958, our committee submitted a report setting forth our conclusions in considerable detail, copies of which have been furnished to Mr. James T. Ramey, Executive Director of the Joint Committee on Atomic Energy.

Our committee recognizes the following major problems associated with the development of a system for disposing of radioactive waste in deep wells:

1. A suitable disposal reservoir must be clearly defined and must be capable of confining radioactive waste solution for indefinite periods.

2. The heat generated by the radioactive material must be safely dissipated.

3. The disposal reservoir must be protected against damage such as fracturing, premature plugging, or other causes.

4. The failure of equipment as a result of corrosion or other causes must be prevented.

5. An economical disposal plant must be designed and safe operating procedure must be developed.

Our committee, on the basis of its consideration of these problems reached the following conclusions:

1. The disposal of radioactive waste from fuel processing is a significant element in the cost of atomic power. While safety is of paramount consideration in the selection of waste disposal methods, cost is the factor that will govern the choice of safe disposal processes. The economic aspects of waste disposal will receive greater attention when atomic energy starts to compete in the market for industrial and domestic power.

2. Radioactive waste can be disposed of safely by injection through deep wells into porous rocks. Obviously, there is a limit to the amount of radioactive waste that can be stored without danger in any particular reservoir. The data required for estimating the safe storage capacity of a particular reservoir can be obtained by laboratory experiments and field tests. It is not possible to state with information now available, whether or not disposal in deep wells will be better than other methods.

3. The solution of the many problems associated with the disposal of radioactive waste in deep wells will present difficulties but no insurmountable obstacles are indicated.

4. Reliable cost estimates cannot be made on the basis of information now available, but preliminary considerations suggest that in some locations, the cost of disposal in deep wells might compare favorably with that of alternate disposal systems.

5. Many of the problems associated with deep-well disposal could be clarified through a program of laboratory work and theoretical study. If such studies confirm the preliminary conclusion that deep-well disposal could be economic under favorable circumstances, the suitability of a particular reservoir chosen for disposal purposes can be tested in the field prior to the injection of radioactive waste.

6. A site suitable for the disposal of radioactive waste should be selected through geological and geophysical exploration. The disposal of radioactive waste in association with secondary recovery or any other oil producing operation is not recommended.

Senator ANDERSON. Someone has sent forward a question: What order of magnitude of elapsed time and money is involved in the proposed test program?

Mr. MOORE. I think there are two parts to such a program. One of them consists of more or less general studies that would be applicable to almost any condition. That would be largely a laboratory and theoretical investigation.

It is difficult to forecast exactly what such a program would involve, but I think it could be a modest program, which would involve several man-year's work and probably would cost a small number of hundred thousand dollars.

Later, in any particular area, work would have to be done that would be specific to that area. That would involve more fieldwork and would be somewhat more expensive.

I would guess that perhaps a cost in the order of a million, more or less, dollars and work extending over 2 or 3 years would be required.

Representative HOLIFIELD (presiding). Mr. Van Zandt?

Representative VAN ZANDT. On page 2 you say here—

> The solution of the many problems associated with the disposal of radioactive waste in deep wells will present difficulties but no insurmountable obstacles are indicated.

What are some of the difficulties?

Mr. MOORE. There is always the possibility of, for example, failure of tubing in some of the wells due to corrosion, leakage, mechanical troubles and things of that kind.

Representative VAN ZANDT. This is tubing that would be inserted in the well if this program were to become a reality?

Mr. MOORE. That is right. There is always a possibility that the formation in which you were injecting the waste might get plugged up. I think after you have injected enough waste you would expect it to become plugged.

I think that the feasibility of the process depends entirely on how much waste you could inject before the formation becomes plugged. I am sure there are going to be some serious problems with the instrumentation to monitor the area to find out where the waste is going. We think that these are problems that can be solved through a program of study.

Representative HOLIFIELD. Isn't it true that you have these wells in many areas of the country and I am thinking now of some arid

areas, probably in Texas, some of the Western States, where oil wells have been drilled down to 10,000, 12,000, or 13,000 feet in some instances and oil and gas have been recovered from those areas to the point of exhaustion, and this leaves substantial area in there which may or may not have been filled with some types of old line salt water, but which is available for the pumping under compression of materials down into that through your pipe.

I know in California, for instance, in my hometown, the gas companies have recently bought a whole zone of the Montebello oilfield there which is an exhausted zone and they are pumping in gas from Texas, using it as storage.

There is another project down in Maryland where the Washington Gas Co. is buying property down there to store gas underground.

I am thinking about these large areas which have been exhausted in a deep well in arid country where this particular strata might be much lower than any of the fresh-water streams, underground streams, that are above it, and why couldn't this liquid waste be pumped into those caverns even though it had to be pumped in under pressure?

You and I both know that you can seal off these pipes by cementing, and they frequently do it at different levels. They seal it off in stratas of oil and gas production and drill to it and go on down to the next point.

Mr. MOORE. That is a possibility and it is one that we considered. Of course, you cannot rule it out. However, in many of these older fields there are a lot of old wells, and we do not know exactly how well these wells were completed.

Representative HOLIFIELD. Sealed off.

Mr. MOORE. Sealed off. There is a possibility of leakage or damage through the old wells and our group thought that it would perhaps be safer if you could find a suitable formation that was not associated with any oil production, which had not been tapped before, and where you would have complete control of everything that was done to that formation.

Senator ANDERSON. Are you connected with the geology work of the company in any way?

Mr. MOORE. Yes, in a general way.

Senator ANDERSON. Are you familiar with the so-called Delaware Basin out in my State?

Mr. MOORE. Generally, yes, sir.

Senator ANDERSON. They have drilled wells there down to about 13,000 feet and I think they were dry most of the way. There was a little oil here and there but most of it was dry. I know of a well where the casing had not been set because they had no production.

Would not that be a place that might lend itself to this sort of thing? I am only trying to find out what you mean by areas of this nature. Here is 13,000 feet. I think the production probably around it is at 10,500. It is several thousand feet below the immediate areas that are producing.

Would it be down far enough to cause no trouble?

Mr. MOORE. I think that is a possibility. As I say, I am not familiar with the details of the geology in that particular well but that is the kind of thing that we were thinking about.

Senator ANDERSON. It would not bother anything else? No oil is in production in or around that area.

Mr. MOORE. I think it would be preferable to put this in an area where there was no oil production or no really good prospects for oil production.

Representative HOSMER. Mr. Moore, it seems to me if you take one of these oilfields which it has been thoroughly drilled and the geology is known, and location, and so forth you can pretty well determine what you could do by way of injection.

You talk in your statement about laboratory experiments and so on and so forth but I cannot detect from the statement or a quick review of this report any illustration of an actual piece of property where the characteristics are well known and theoretically applying this.

Mr. MOORE. Well, there are several formations in the gulf coast that we know about that would be suitable for this if it were not for the association with the oil production. There are quite a number all over the country.

Representative HOSMER. I am talking just about specifically picking out some area where the geology is well known and working the problem out. You left us with so many questions to be answered. I was wondering if we couldn't get some in that way.

Mr. MOORE. Yes, I think one of the recommendations in our report was that a thorough study be made of all of the sedimentary basins in the United States with a view of picking particular areas.

One area that intrigued some of us who were on this committee was the Atlantic coastal plain.

Senator ANDERSON. You may know of a few people who have unsuccessfully tried to hit something along the coast of the Carolinas.

Do you know that it is pretty well believed that there is no oil south of the Colorado River in Texas? At least I tried to find it a few times and couldn't.

Would you choose an area like that where the wells are not very deep; would you go over to the Delaware Basin where they are extremely deep, or would you go to the Carolinas practically anywhere you want to and go down a couple of thousand feet where nothing has ever been discovered?

What kind of an area are you looking for?

Mr. MOORE. We drilled a couple of wells on the Carolina coast about 9,000 feet deep. Down at the bottom of those wells there was a very thick section of porous, very permeable, sand that would have a lot of the characteristics that we think are desirable for this kind of thing.

I really hate to pinpoint an area because it is going to take some detailed geological study, but I am just trying to cite an example of a particular area that might be suitable.

Senator ANDERSON. You would stay away, for example, from areas in Mississippi that still seem to have some possibilities and have not been drilled deep? You might have a thousand-foot well along the Atlantic coast that is reasonably deep, but you can find many areas where that is not true.

You would go into some area like that, in general? I am not trying to pinpoint it either because that is hard for you to do, but I am just trying to find the type of area that you prefer.

You would choose the Carolina coast as against something in the tidelands areas of Texas and Louisiana that would be deep but not productive?

Mr. MOORE. We would prefer that for the first operation. After we knew something about it, we might find out that we can operate in such areas as, say, the Texas Gulf Coast, or some of the sand areas of west Texas, but in order to be absolutely safe, we would prefer to stay away from the oil-producing territories at first.

We might later learn that we can do it.

Representative HOSMER. What do you calculate the quantity of waste would be that you would be putting down in one of these wells? You get 20,000 barrels a day down in a good injection well and I think a lot more than that in more modern ones, so you are going to run out of material pretty fast at that rate.

Mr. MOORE. Maybe we wouldn't because we recommend that the relatively high-level waste be diluted.

Representative HOSMER. Yes.

Mr. MOORE. So it would depend entirely on your factor of dilution. Now, I would think that we would want a well that would take a few thousand barrels a day of total fluid and the concentration of high-level waste that you would put in that I think would depend on the particular plant, the nature of the waste, the specific circumstances that prevail.

Representative HOSMER. You want to cut down your heat concentration, too.

Mr. MOORE. That is right.

Representative HOSMER. Still we are not talking in any amounts of an injection like you are talking about when you are repressuring oil fields.

Mr. MOORE. No.

Representative HOSMER. This is just a peanuts operation, is it not?

Mr. MOORE. Well, we got the impression that some of these large fuel procesing plants might have quite a large quantity of high-level waste associated with it and to dilute that down to a concentration which we would feel reasonably safe in handling would probably mean a significant rate of injection.

As I say, we are thinking, guessing, something of the order of a few thousand barrels a day of total fluid.

Representative HOSMER. You are probably not going to have more than a barrel a day to put in.

Mr. MOORE. I thought we probably would.

Representative HOLIFIELD. 52 million gallons are now in storage at Hanford.

Representative HOSMER. Take that number in barrels and divide it by the number of days a year and you would probably find you would be putting all that stuff down.

Mr. MOORE. That is a little over a million barrels and that is fairly high-level stuff.

Representative HOLIFIELD. It is high level.

Mr. MOORE. Diluted by a factor of 20 or 50, that would be 20, 50 million barrels you would have to dispose of and at, say, a thousand barrels a day, that would be, oh, a 15-year operation.

Representative HOSMER. You are not going to get any thousand barrels a day.

Mr. MOORE. Sir?

Representative HOSMER. I mean you can use wells that would put down a great deal more than that.

Mr. MOORE. Oh, yes, certainly you could.

Representative VAN ZANDT. I notice on page 2 there, you say in subparagraph 2:

> Radioactive waste can be disposed of safely by injection through deep wells into porous rocks.

Using our imagination, would it be possible to find the type of rock and then drill wells that would be competitive with existing disposal facilities from a cost standpoint?

Mr. MOORE. I think so. I think in general, there is enough known of the general geology of the country to be able to say generally where these locations are likely to be found.

Representative VAN ZANDT. With your knowledge of geology, would you say that it would be possible to establish we will say, disposal areas, geographically throughout the United States to meet the needs of the atomic industry as it develops?

Mr. MOORE. Once the practicability of this had been tested and a little bit more specific information had been developed relative to what was involved in the operation, I think that that could be done.

Representative VAN ZANDT. In other words, we would cut down the transportation of this waste material which has a certain amount of radiation that could be disastrous if it become involved in an accident?

Mr. MOORE. Yes.

Representative VAN ZANDT. Being from Pennsylvania I am just wondering what value the many abandoned oil wells in Pennsylvania may have in this program.

Senator ANDERSON. They might be pretty shallow.

Mr. MOORE. They would be shallow and a great many of those wells were drilled a long time ago but the records are not too good on many of those wells.

I would hesitate to suggest that you put them in old fields of that kind.

Representative VAN ZANDT. A moment ago you indicated that you would not want to develop a disposal area in, or adjacent to, an oil field. Is that for fear of contamination of the oil field itself?

Mr. MOORE. For fear of contamination of the oilfield and also, because of the possibility of leakage through these old drill holes that might not have been properly cemented and properly abandoned.

Representative VAN ZANDT. Using our imagination, can you describe what a disposal area might look like in years ahead after the R. & D. effort has been completed?

Mr. MOORE. Yes. I would imagine that there would first be a plant for chemical processing, and filtration of the high-level wastes. We may have to store it for some time to let it decay a little bit.

The waste might have to be filtered. There may be some chemical treatment involved to make it compatible with the water so that you didn't precipitate something from the water that would plug the formation.

Then I think that associated with this would be some wells in the same formation, same reservoir, in which you are going to dispose of the waste that would take water out of that formation in order to lower the pressure.

By lowering the pressure in the formation in which you are going to inject the waste you could fix things so that if any leakage occurred it could be leakage from the outside into the contaminated area rather than from the contaminated area out.

I think we would want to conduct an operation that would involve some ordinary, normal producing wells for producing salt water from the area to be contaminated. Then there would be a plant with the necessary pumps and equipment with two or three injection wells. These would be the wells into which the waste would be pumped. These wells would have to have proper shielding. The piping would all have to be shielded. These wells would have a lot of special alloy pipe in them, things of that kind.

Then there would still be some other wells around that would be used for instrumentation and monitoring purposes, so you could trace the waste as it went out, and you could get some kind of check on where the waste is going.

We think that the area involved might be, oh, 50,000 to 100,000 acres. It would not be—certainly shouldn't be—land which was valuable from the surface. If, for example, some coastal location were possible, I am sure there would be a lot of swampland or even bay area that could be used for the purpose.

Senator ANDERSON. You said put down pipe that can take care of this high radiation. Do I understand that you are going to use abandoned fields that still have pipe in them, and then try to put down new pipe with shielding in it?

You know, it is hard enough to handle the pipe in existing wells without putting down a lead pipe. How would you handle 10,000 feet of lead pipe?

Mr. MOORE. The shielding I was thinking of would be at the surface, piping from the plants to the pumps and things of that kind.

Representative VAN ZANDT. What would you do with the salt water?

Mr. MOORE. The salt water that would be produced would be handled two or three ways. You would use part of it to dilute the waste with. The excess could be reinjected into another porous sand body, that would be, say, above the disposal area.

Representative VAN ZANDT. This area would be posted, would it?

Mr. MOORE. I certainly think it should be.

Representative HOLIFIELD. The point you brought up about using an old field is a very valid point because in most of these old fields there are improper records of whether the pipe was cemented off at different levels and whether it was taken out or lost in the hole and all that sort of thing.

I lived in the middle of the oil field in California. In fact, I have an abandoned oil well on my property. It was drilled before I bought the property and I have no oil interest in it, sorry to say, but no one knows the history of these wells in any development field.

Wildcatters and stock companies and so forth went in and they had very irregular operations. Now, on your big leases, like your Standard leases and Shell and a few of those big companies, they have pretty complete records of each hole. Where these little second wells were drilled, wildcatter wells, there were very poor records.

You put that down one place and it might be coming up in another.

Are there any further questions?

Representative HOSMER. I want to ask has any thought been given to grouting this stuff into suitable formations?

Mr. MOORE. Grouting the waste?

Representative HOSMER. Yes.

Mr. MOORE. Yes. The Continental Oil Co., has considered that and some of the engineers with that company I think, feel that a practical method could be developed of making some kind of a slurry that will set into a kind of cement and injecting it through fractures created in a relatively impervious formation, such as shale.

Representative HOSMER. Thank you.

Chairman DURHAM. Did your committee try to assess or determine how much storage is available in this kind of a method in being at the present time, or is that impossible?

Mr. Moorse. I am sure that the availability of the storage is so large that it would be tremendous in comparison with any amount of——

Chairman Durham. Would you hazard a guess as to how much is available if this thing does work out?

Mr. MOORE. I would say many hundreds of times the requirement for, say, 100 years or so, but the problem there is while the total capacity is available, it may not always be available at the exact spot that you want it. It is not available all over.

Chairman DURHAM. I realize that, but I think you are right in stating here that it would have to be a pretty active determination of the geophysical and the geological strata and everything else involved in such an operation.

That would have to be done by drilling outside your hole.

Mr. MOORE. It would be, yes.

Representative HOLIFIELD. There would be something done toward locating these processing plants in areas near to a disposal area in the future. Of course, we are hooked with some of them now like at Hanford. It would be very difficult to take that 52 million gallons and transport that material to a permanent place.

Mr. MOORE. I think that might be one of the main merits of a disposal system of this kind. It might give you a little bit more flexibility in locating your plant next to a disposal system.

The system that Mr. Heroy described this morning of using salt mines, I am sure is a very fine system, but if there were other compelling reasons for locating plants outside of areas in which these salt mines were available this might have some merit.

Representative HOLIFIELD. Considering the expense that we are going to now in building these steel tanks and concrete canyons and so forth, to handle this material, I think it behooves us to get on with the job of finding out what we can do about this as quickly as possible because it is obvious, when you are paying $2.60 to $4.90 a gallon to dispose of this, that you are going to affect the effects of power production very sharply and the Government cannot keep on absorbing this if we are going to have a true picture of the economics of atomic power.

Are there any further questions of Mr. Moore?

If not, thank you very much, sir, for your testimony.

(The formal statement of Mr. Moore, in the form of a report by the Subcommittee on Disposal of Radioactive Waste of the American Petroleum Institute follows:)

PROBLEMS IN THE DISPOSAL OF RADIOACTIVE WASTE IN DEEP WELLS

A Report prepared by the
Subcommittee on Disposal of Radioactive Waste
Central Committee on Drilling and Production Practice
Division of Production
AMERICAN PETROLEUM INSTITUTE

SUBCOMMITTEE ON DISPOSAL OF RADIOACTIVE WASTE

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SUMMARY

Among the major problems in a system for disposing of radioactive waste in deep wells are (1) containment of the waste in suitable reservoirs, (2) dissipation of the heat generated by radioactivity, (3) protection of the disposal reservoir against damage by fracturing or plugging, (4) corrosion. The subcommittee submitting this report is of the opinion that all of these problems can be solved and that a safe deep well disposal system for certain kinds of waste can be developed.

As the technology of atomic energy advances, the merits of various processes for waste disposal will be determined by economic considerations. Sufficient data are not available to permit good cost estimates to be made, but preliminary estimates suggest that, for favorable locations, deep well disposal might be one of the more economical means for handling waste. For this reason, further study of the process may be justified.

This report outlines several investigations which the subcommittee believes would be important elements of such a study.

I.

THE BACKGROUND OF THE A.P.I. SUBCOMMITTEE

This report on the problems associated with the disposal of radioactive waste in deep wells was prepared by the A.P.I. Subcommittee on the Disposal of Radioactive Waste on the basis of investigations extending over a period of several months. This subcommittee was established on July 3, 1957, by direction of the Executive Committee on Drilling and Production Practice (this committee is one of the major committees which plans the activities of the Division of Production of the American Petroleum Institute), as a result of a request by representatives of the Atomic Energy Commission for the assistance of the petroleum industry in studying the waste disposal problem. The subcommittee was established without specifically defining the nature and scope of its assignment, and, accordingly, the subcommittee itself had to determine its objectives during the course of discussions with representatives of the Atomic Energy Commission and other interested organizations. Before preparing this report, the subcommittee held the following meetings:

1. At Chicago on November 10, 1957, for organization.
2. At Oak Ridge on January 21-24, 1958, for discussions with the staff of the Oak Ridge National Laboratory.
3. At Dallas, February 6, to decide on scope and plan for Committee activity.

4. At Hanford from March 31 to April 4 for similar discussions with the staff of the Hanford Atomic Works.

5. At Colorado Springs, May 7-8, to prepare preliminary outline of subcommittee report.

6. At Kilgore from May 19 to May 23 for a review of technical problems with members of the staff of the Atomic Energy Commission, the Oak Ridge National Laboratories, the Hanford Atomic Works, and the U. S. Geological Survey, and to prepare a preliminary draft of this report.

7. At Houston, on October 7, 1958, to revise the preliminary draft on the basis of the helpful comments and suggestions received from several sources.

The subcommittee also arranged for a visit of the members of the staff of several organizations interested in the problem of radioactive waste to oil field installations and research facilities of several major oil companies and service companies in Texas and Oklahoma during the period May 12 to May 20, 1958.

II.

OBJECTIVES OF SUBCOMMITTEE

Storage in porous rocks at depth is only one of several methods for handling radioactive waste. It is probable that, as the technology of atomic power develops, many kinds of disposal systems will be used, the choice of specific systems depending in part on the nature of the waste and the locations of the processing plants. Ultimately, the economics of the disposal process will have to be taken into account, and preliminary considerations indicate that, if deep well disposal can be used at all, the associated costs will compare favorably with the costs of other methods. This is a major reason for careful consideration of the feasibility of deep well disposal.

In planning its activities the A.P.I. subcommittee has attempted to reach four major objectives:

1. To review, with the experience of the petroleum industry as a background, the problems of deep well disposal and to arrive at some tentative conclusions as to the probability of successfully solving these problems. In this the subcommittee confined its investigation to the practical and economic considerations of disposal in deep wells, and did not attempt to make comparisons with or evaluation of other disposal methods.

2. To call to the attention of the Atomic Energy Commission and their contractors those parts of the technology of the petroleum industry which might be useful in the development of a safe and economical process of deep well disposal.

3. To suggest specific approaches to the problem which might help to guide research, development, and pilot operations. In doing this the subcommittee presents some very rough preliminary figures indicating the order of magnitude of the cost which might be involved in deep well operations.

4. To indicate to the A.E.C. and their contractors some studies which the subcommittee felt could be profitably undertaken in the development of a deep well disposal process. Most of these have been previously suggested by others, and some are under active investigation. They are listed here in order to give some feeling for the magnitude of the research and development program that might be associated with the development of the deep well disposal system.

III.

PROBLEMS IN DEEP WELL DISPOSAL

In considering the technical problems, the subcommittee has concluded that an aquifer* for disposal purposes must be deep enough to preclude any possibility of contamination of potable ground water or the surface of the earth. In some areas the required depth may be fairly shallow; in others it may be quite deep. However, it is probable that at the first deep well disposal site the aquifer should be deeper than 3,000 feet.

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*The term "aquifer" is used by petroleum engineers in its broad sense to describe water bearing geologic strata. Some hydrologists use the term in a more restricted way to refer to shallow beds which are sources of fresh water. To avoid confusion, some members of the staff of the United States Geological Survey have used the term "salaquifer" to define porous and permeable beds or formations situated several thousand feet or deeper in the earth which are saturated with salt water and which lie below the potable water zone. Thus, the term "salaquifer" describes the kind of reservoir which the subcommittee has considered for waste disposal. However, in preparing this report, the subcommittee is reluctant to adopt terms not yet in general use, and it will use the term "aquifer" in its broad definition.

The principal problems to be solved in the development of a safe method of deep well disposal are the following:

1. The disposal reservoir must be clearly defined and must be capable of confining radioactive waste solution for indefinite periods.

2. The heat generated by the radioactive material must be dissipated at a sufficient rate to prevent excessive increase in temperature.

3. The disposal formation must be protected against damage such as fracturing, premature plugging, or other causes.

4. The failure of disposal well equipment as a result of corrosion or other causes must be prevented.

5. An economical disposal plant must be designed and safe operating procedure must be developed.

IV.

FLUID CONTAINMENT

The success of any system for disposing of radioactive waste in deep wells depends upon the ability to confine the waste in a suitable formation of porous rock for indefinite periods of time. Of the many kinds of porous geological formations that are known, there are some that appear definitely unsuitable because of possible communication with the surface or with other porous beds that contain potable ground water or other valuable minerals. Nevertheless, there are tremendous areas in which geological formations which would provide suitable reservoirs for the storage of radioactive waste might be found.

The kind of rock which would be most favorable for waste storage would depend upon the chemical nature of the waste. Most raw high-level waste from the present processing plants is strongly acid, and it is probable that porous limestone would not be suitable for the storage of an acid waste. There are also other reasons for questioning the suitability of limestone reservoirs for waste disposal. First, porous limestone beds, lenses, and reefs tend to be developed around the margins of sedimentary basins; basin margins are also major areas of petroleum exploration and production. Second, fracturing and the presence of channels characteristic of many limestone reservoirs would greatly complicate the problems of adequate containment and monitoring of the injected fluids. Third, the continuity and uniformity of porosity in a limestone are generally uncertain.

Thick shale formations in which fractures exist or can be created are a second type of rock which could be used for storage of fluids. The problems of containment and monitoring would be essentially the same in a fractured shale as in a fractured limestone. Further, the storage capacity in a fractured shale would usually be less than in other kinds of rock.

These considerations tend to focus attention on sandstones or beds of unconsolidated sand as suitable reservoirs for waste disposal, although it is not possible in any preliminary consideration of the problem to rule out any class of porous rock. One type of structure which appears favorable for waste disposal is the closed sand lens bounded on top and bottom by nearly impervious rock such as shale. Such a lens might at one time have been an offshore sand bar or a segment of a river channel. Obviously, a considerable number of wells would be required to establish the fact that any given sand body was lenticular and to delineate its boundaries. Upon finding such a lens at a suitable location, two additional problems would arise; the first of these would concern the isolation of the lens from other permeable strata. This could be investigated by a suitable test program involving the production of water from the lens and the measurements of pressures over a suitable period of time. The second problem would concern the ability to maintain the hydraulic isolation of the lens over the required period during which the waste is to be stored. This would depend to a considerable degree on the manner in which the disposal operations were carried out and on the adequate surveillance of the site even after disposal operations had been discontinued. It would be necessary in such a formation to withdraw a volume of water equal to or greater than the volume of the injected waste. Suitable measures would have to be taken to prevent the generation of excessive pressures due to any heat developed by the radioactive material.

While closed sand lenses could probably provide suitable waste disposal locations, there are certain advantages to extensive aquifers for waste disposal. These are found at depths of several thousand feet over wide areas in the United States. They consist of beds of porous rock ranging from a few feet to several hundred feet in thickness. A typical aquifer suitable for waste disposal might consist of a series of inter-bedded sand and shale with a thickness of one or two hundred feet or more. Its areal extent might be many thousand square miles. It would be bounded on top by a layer of impervious rock of substantial thickness. The staff of the Oak Ridge National Laboratory has pointed out the advantage of a thick formation of rock salt to provide such a cover, although a thick section of shale would also be satisfactory. Petroleum technologists have made extensive studies of the movement of fluids in such aquifers, and they have developed a good understanding of the relation between the rate of fluid movement, the permeability of the rock, and the pressure distribution in the aquifer. With data obtainable from well logs, analyses of core samples, and data on wells as fluid is produced from or injected into the aquifer, the fluid movement can be predicted with acceptable accuracy. Furthermore,

by producing or injecting fluids at selected points, the fluid movement is controllable. Any reservoir of this kind could be subjected to thorough testing to provide assurance that the fluids could be controlled and that the injection operations would not induce fracturing of the rock which might lead to uncontrolled migration of the waste. By providing wells at suitable distances from the injection well in which measurements of radioactivity could be made, the advance of the waste solutions could be monitored and any serious deviation from the predicted path of movement could be observed and controlled.

To insure confinement of the waste in the area selected, it might be advisable to create an area of low pressure around the disposal wells by producing water at a rate somewhat in excess of the rate of waste injection from a few bleed wells surrounding the disposal site. Under such circumstances, the fluid movement would tend to be a movement of water into the zone selected for waste disposal rather than the leakage of waste out of the zone.

It is recognized that the confinement of waste in deep reservoirs presents many difficult and serious problems. However, it is believed that there are extensive areas in which suitable reservoirs for storage of the waste can be found, that the suitability of a reservoir can be adequately tested before the injection of radioactive material, and that with a properly designed and operated system, the waste can be safely confined and retained.

V.

HEAT DISSIPATION

There is some concern that heat from strong radioactive waste may result in exorbitant temperature rise in the aquifer, with accompanying danger of damage to the disposal operation. Fortunately, it is possible to estimate, by computational methods now used in the petroleum industry, the temperature rise to be expected. Control of the maximum temperature may be achieved by storage of the waste for some time prior to injection into the aquifer or by dilution of the waste. Even though the dilution may result in relatively large volumes of waste, the operation should present no formidable problems. In some existing salt water disposal operations, for example, as much as 20,000 barrels of salt water are injected per day in a single injection well, although such high injection rates are attained only in thick sections of high permeability.

With regard to the question of the temperature permissible in the aquifer, a requirement that the liquids be maintained below the boiling point would undoubtedly set a safe limit. This temperature is computable from the hydrostatic pressure expected at a particular depth. At a depth of 5,000 feet, for example, the hydrostatic pressure in the aquifer

is normally about 2300 psi and the earth temperature usually somewhat less than 200° F. The vapor pressure of water approaches this hydrostatic pressure only when the temperature reaches about 700° F. Arbitrarily, then, the temperature desirable might be taken as that which will yield a vapor pressure of, say, 2000 psi. This temperature is in the vicinity of 630° F, which would permit a temperature rise of more than 430° F. Selection of a temperature in this range would, under the assumed conditions, provide an ample margin of safety. Indeed, unless the waste is confined in a closed lens, vaporization of water should not be particularly harmful. There is a possibility that some endothermic reactions involving reservoir materials might take place as temperatures in the aquifer approach about 500° F, which would result in the absorbtion of heat. High temperatures per se should not be damaging to the aquifer. Evidence obtained during the course of in situ combustion operations for oil recovery indicates that temperatures as high as 1400° F. have been experienced in a producing oil sand with no significant damage to the sand structure.

Laboratory investigations carried out by the petroleum industry in connection with water flooding operations have shown that ions foreign to the interstitial water are usually adsorbed on surfaces of the rock particles. Some fixation occurs even in comparatively clean sands, and the amount sorbed may become quite large if the rock contains much clay or silt. Some consideration has been given to the possibility that, in a dirty or shaly sand, fixation of radioactive elements by adsorption in the vicinity of an injection well might result in an unduly high concentration of radioactivity with production of a localized thermal high. Where several radioactive elements are present, as in the case of actual waste, there is the further possibility of a chromatographic separation, concentrating particular radioactive elements in annular rings and causing zones of high temperatures. Generally, the experience of the petroleum industry in drilling mud technology and in well rejuvenation indicates that adsorption of most metallic ions decreases with increasing temperature. In the event of a localized buildup of radioactive elements by adsorption, the consequent generation of heat would serve to desorb some of the radioactive material, thus reducing the rate of localized heat generation. Inasmuch as the fixation of the radioactive ions serves to confine positively a part of the waste, there is a considerable benefit to be derived from adsorption of waste in proper amount.

An important point to be made here is that, either by storage prior to injection or by dilution, the specific radioactivity of the waste going to the disposal wells can be governed, and the maximum temperature developed can be controlled by proper design and operation of the disposal system. Certainly the generation of heat may be one of the factors which limits the capacity of a particular disposal plant, but this would affect the economics, rather than the ultimate safety, of the operation.

VI.

DAMAGE TO AQUIFER

A deep formation used for disposal purposes may be damaged from either or both of two principal causes:

1. Fracturing of the formation.

2. Loss of permeability due to plugging.

If the pressure of the injected fluid increases above some critical value, the formation will rupture, allowing irregular and uncontrolled flow through the resulting fractures. Experience in the petroleum industry shows that damage of this kind will probably be confined to the disposal reservoir and is unlikely to result in the rupture of the overlying rocks. Subsequent reduction of pressure will usually result in closing of the fractures so that damage is not permanent. Nevertheless, fracturing the kind of formation suitable for a disposal aquifer should be avoided in order to maintain proper control over the movement of the injected fluids.

It is easy to detect the occurrence of fracturing since it is accompanied by a great increase of the rate of injection without a corresponding increase in pressure. In some cases injection pressures actually decrease after initial fracture. The pressure at which a formation will fracture and accept fluids is primarily dependent upon the stress conditions in the rock. Most formations will fracture when fluid pressure in the range of 0.6 to 1.0 psi per ft. of depth is exerted against the formation. Pressures approximating 1.0 psi per ft. of depth are thought to be necessary to create horizontal fractures, but much lower pressure may produce vertical fractures. Neglecting friction losses, surface injection pressures of from one-fourth to one-half psi per ft. of depth would be necessary to cause fracturing. The disposal operation considered here does not contemplate the injection of waste at pressures nearly this high. It would be a comparatively simple matter to test the disposal formation at any particular location for susceptibility to fracturing by injecting water at moderate pressures.

Plugging of the formation will be a far more serious cause of damage. Experience of the petroleum industry in water injection systems shows that in general there are two types of plugging: that which plugs the pores in the main body of the sand and that which results from the formation of an impermeable crust at the sand face. Plugging in the body of the sand in oil field practice usually occurs as a result of the swelling of the clay minerals in the rock. Plugging at the face of the sand in the well bore may result from a number of causes, among which the following are important:

1. Injection of fluids containing suspended solids, which are often mineral precipitates.

2. Precipitation as a result of the comingling of the injected water with water of incompatible composition.

3. Accumulation of corrosion products.

4. Precipitation resulting from the action of micro-organisms, of which sulphate reducing bacteria are perhaps the most important.

5. The accumulation of tar-like material as a result of the incomplete separation of oil and water at the surface.

While cleaning out wells to improve injectivity is a common oil field practice, these operations will be complicated and hazardous if the well and formation have been used for the injection of radioactive waste. Therefore, it will be desirable to treat the radioactive waste to minimize plugging. Likewise, it will be desirable to provide for the separate treatment of any water used as a diluent for the radioactive solutions.

The physical and chemical characteristics of the waste to be disposed of at a particular plant cannot be foreseen. Consequently, only general suggestions can be made concerning the treatment that may be required to control precipitation or plugging. Filtration will be required to remove any suspended solids, but other treatment may also be desirable. It may be desirable to use some additives such as corrosion inhibitors, chelating agents or wetting agents for specific purposes. The effects of heat and ionizing radiation on chemicals used for waste treatment will present problems not encountered in oil field practice.

Although the composition of the waste is not known, it is highly probable that it will be an acid solution which will require dilution to a considerable extent. It is unlikely that the waste will have been mixed with any substantial amount of oil so that plugging from tar is not likely. The results of research on the effects of ionizing radiation on micro-organisms shows that the radiation destroys most, but not all, bacteria which may get into the waste, so that although there is a possibility that some trouble may be experienced as a result of microbial activity, it is unlikely to be as serious as it is with common oil field waters. Strongly acid solutions result in shrinking rather than the swelling of most of the clay minerals that are likely to be encountered, so that it is not anticipated that the injection of acid waste will result in trouble from this cause. The accumulation of corrosion products is a potential source of trouble, but the control of corrosion will be dealt with in a subsequent section.

The remaining important problem appears to be precipitation due to the incompatibility of the injected fluid with the disposal reservoir. Some waste is very likely to contain elements such as aluminum or iron, which tend to form gelatinous precipitates at moderately high pH. Even though the waste is injected at a very low pH, the adsorption of hydrogen ion by clay minerals, reactions with carbonate minerals in the rock, or by dilution with connate water, will result in increase in the pH. At first glance this appears to present a very serious problem, but there are data which indicate that the reduction in permeability due to these effects might not be as serious as preliminary considerations would indicate. The mechanism of displacement of interstitial water by the injected waste may be such that the precipitation may be delayed or prevented, or the precipitates may be formed in parts of the pore network which are out of the main channels of flow. While plugging in the body of the sand by precipitation is a serious problem, it is susceptible to study and might be controlled by suitable treatment of the waste.

In oil field work, plugging can be alleviated by backwashing, chemical treatment or working over the well. These operations will certainly be much more difficult in wells used for the disposal of radioactive waste than in those used for water injection, but there is a reasonable probability that suitable techniques can be developed for restoring the injective capacity of wells which have been used for radioactive waste disposal. If, however, this proves to be impossible, the damaged well must be abandoned and a new one drilled. The evaluation of this aspect of the deep well disposal system is accordingly largely an economic problem and the merit of the system is determined by the amount of waste which can be injected before abandonment becomes necessary. It is probable that data sufficiently accurate for estimating cost approximately can be obtained by laboratory studies and pilot operations. The performance of the disposal wells and the disposal aquifer will have to be tested finally in the field.

VII.

CORROSION

The injected waste will undoubtedly be highly corrosive and it will be necessary to use stainless steels or non-ferrous metals for some of the tubular equipment in the disposal wells. The Atomic Energy Commission and its contractors have extensive experience with corrosion in fuel processing plants which would permit them to select suitable alloys and to determine the extent to which they should be used. Basically, the disposal well will consist of several concentric strings of pipe. The tubing through which the radioactive waste is pumped would unquestionably have to be of corrosion resistant alloy, but it may be that with proper protective measures a suitable grade of standard oil field tubing or casing could be used for the other strings. For limited special applications, non-metallic pipe made from glass, reinforced plastic, asbestos cement or other material

might be used, but the serviceability of this material under the conditions of temperature and radiation to be expected would have to be tested.

Measures to prevent the corrosion of steel pipe would include protective coatings, inhibitors, cathodic protection, or a combination of these methods. In developing protective measures, it would be necessary to determine the effect of temperature and radiation on organic coatings or inhibitors which might be considered for use. A particularly bad corrosion problem is presented by strongly acid solutions containing both chloride and nitrate ions. This would make the problem of corrosion control particularly difficult with waste from processes which use aqua regia to dissolve spent fuel elements. If, however, the waste is derived from a plant which uses nitric acid, the problem would be simpler. In such a plant it would probably be inadvisable to dilute the waste with salt water at the surface in order to avoid comingling chlorides and nitrates. If other considerations make it desirable to dilute a nitric acid waste with salt water, this could probably be accomplished without serious trouble by providing separate conduits in the disposal well for the waste and the diluent, allowing the mixing to take place at the bottom of the well below any steel pipe.

External corrosion of steel casing occasionally presents a problem in oil wells: With the casing properly cemented to the surface, external corrosion should not present any difficulty, but if trouble from this source is suspected, the condition can usually be controlled by cathodic protection.

The detection of pipe failure due to corrosion can be readily detected by careful recording of pressure and flow rate in the tubing and casing strings, or by radioactivity logs run in the disposal well after displacing the radioactive fluid. However, it would probably be desirable to provide monitoring wells close to the disposal well in which radioactivity logs could be run or from which samples of water could be obtained at frequent intervals to detect leakage in the event of failure of all pipe strings in the disposal well in a manner which would allow the escape of radioactive solution to formations above the disposal aquifer.

In conclusion, corrosion will undoubtedly present a serious problem in any system of disposing of radioactive waste in deep wells, and appropriate pilot plant and field research work should be done to develop techniques for its control. Undoubtedly, satisfactory measures can be developed. The main question is the economic one of the cost of adequate protection.

VIII.

WELL DESIGN AND OPERATION

Among the primary considerations in the design of a well, through which it is proposed to dispose of radioactive waste, are the quantity and composition of the liquid waste, and the physical and chemical changes

that may take place. These determine the amount of diluent, if any, to be used, and the measures required for corrosion control. There are, of course, many other design variables and some will be discussed.

The disposal well should be completed in a porous and permeable formation below any potable water zone; these formations will contain water, of various degrees of salinity. It would be helpful if the disposal formation were competent rock which would allow a "barefoot" completion, that is, one in which no pipe, liners, or other equipment need be placed opposite the disposal horizon. For the injection well, the best oil field practice is suggested with more rigid specifications included for some of the important equipment and completion practices.

By way of example, suppose a disposal well of a depth of 5,000 feet below the surface is to be planned: at this depth, bottom hole temperatures somewhat less than 200° F. can be expected and pressures of the magnitude of 2300 psi will probably be found. The casing program and cementing practice will be suggested with these data as a basis.

A well drilled to 5,000 feet would be designed so that the first outer string, or "surface string" of casing would be set below any fresh-water strata (1,000 to 1,500 feet below the surface) and encased in cement for its entire length to protect the upper strata of fresh water. The main string of pipe, in an experimental or test well which might later be converted to disposal service, could be a standard size of seamless steel casing of approximately 9-5/8 inches outside diameter, set in a drill hole of sufficient diameter to permit at least one inch of cement to be placed outside the pipe for its entire length. The quality and wall thickness of the pipe, based on a suitable factor of safety could be determined by API standard for bursting pressure, tension, and collapse generally employed. Casing centralizers, wall scratchers, stage-cementing tools, and the kind of cement used should follow the most advanced practices of the oil industry. To minimize corrosion by earth electrolysis, suitable cathodic protection of the well might be provided.

There may be several important advantages to a system which would keep the waste and the required diluent separate while flowing through the disposal well, allowing them to mix in the open hole below the casing seat. Of course, this would be advisable only if the composition of the waste and diluent were such that no precipitates would form as a result. In such a system the waste would be injected through a small diameter, perhaps 1-1/2 inches, alloy tubing, and the diluent through the annulus between this string and the 9-5/8-inch steel casing. Some benefits of such a system would be:

1. Cooling of the steel casing and the cement behind it, thus reducing the danger of damage as a result of thermal stresses or of deterioration of the cement or the rocks outside the cement.

2. Reducing the size of the string of special alloy tubing required to handle highly corrosive waste.

3. As previously mentioned, preventing mixture of acid nitrate solutions with chloride solutions, if salt water is used to dilute nitrate waste.

4. Improving the chance that some system might be worked out to decontaminate the tubing sufficiently to remove it for replacement if this should be necessary.

5. Proper control of a properly designed well would permit the operation of the system in such a way that at any level in the well, the pressure of the salt water would be higher than that of the waste, so that in the event of a minor leak in the small tubing, the water would flow into the waste stream, instead of the waste escaping into the salt water confined in the steel casing.

The alloy tubing carrying the waste should be welded, rather than threaded. If it should be desirable to remove the tubing, there might be a chance of decontaminating it by thorough washing, pumping the wash solution into the disposal aquifer, followed by pumping expendable scrapers through the tubing to remove the contaminated surface layer of metal.

A disposal well with a single string of tubing inside the casing would have the great advantage of mechanical simplicity, but it would probably not be practical to run instruments in such a well to provide information on radiation, temperature, or pressure. These data would have to be obtained, if necessary, in a nearby well drilled for the purpose. On the other hand, a more complicated well head would permit running a second string of tubing alongside the alloy string, through which instruments could be periodically run or which could carry the leads to instruments permanently set near the bottom of the well. The second tubing string might also be useful in well repair operations. Such tubing should be at least 2 inches, and preferably 2-1/2 inches in size.

The well head assembly would be as simple as possible and below ground level, properly protected and shielded against any possible harmful radiation. The fluids entering the well should be regulated from a remote location and provisions made for adequate control. Adequate protection should be provided against radiation from the flow lines, treating plant, filters, and storage tanks. Instruments should be provided to obtain essential data such as rate of flow, temperature, pressure, and specific gravity of the waste and diluent. If it is possible to operate the disposal system so that the waste and diluent flow into the aquifer by gravity, while maintaining a vacuum at the well head, it would be desirable to record the fluid level in the alloy tubing and the annular space

by echo sounding devices such as those used to measure fluid levels in pumping wells. Fig. 1 shows a suggested cross section for a disposal well.

The fluids injected into the disposal well should be chemically stable and free of plugging materials. In the suggested plan herein, it is desirable that the waste fluids flow into the disposal well and thence into the aquifer, without the necessity of pump pressure. In order to maintain this type of flow over a long period of time, the formation or strata into which the fluids are to be injected must be of a relatively high permeability, the fluids injected must be chemically stable, free from suspended matter, and compatible with the fluids in the aquifer, and the pressure in the aquifer must be less than the hydrostatic head of the column of injected fluids.

Experience shows that some plugging of the bore hole at the bottom of the well will take place and the ideal rate of flow cannot be indefinitely maintained. Therefore, an effort should be made to design the disposal well so as to provide, from the beginning, the best opportunity for cleaning the injection well. Over the years, the oil industry has devised many ingenious means and methods for working at great depths inside a bore hole or well casing. The unknown factor of radioactivity will require modification and adaptation of these methods. Nevertheless, the repair of disposal wells is still within the range of possibility and practicability, and means for accomplishing this should be worked out when the occasion demands.

In the meantime, the abandonment of the well must be kept in mind; after all, it is a matter of economics whether a well, be it an oil well, salt-water disposal well, or radioactive waste disposal well, should be repaired or replaced with a new well. The problem is met quite frequently in the oil producing business. Well repair and "fishing jobs" are expensive undertakings at the best - the estimated cost of repair must be compared to the cost of drilling a new well. In any case, it would probably be desirable to provide an alternate injection well, as a stand-by, in case it becomes necessary to take a disposal well out of service.

The disposal of radioactive waste through deep wells to an underground or subsurface reservoir or aquifer will present different and complicated problems, but not insurmountable obstacles. The advisability of handling waste in this way will be determined primarily by economic considerations.

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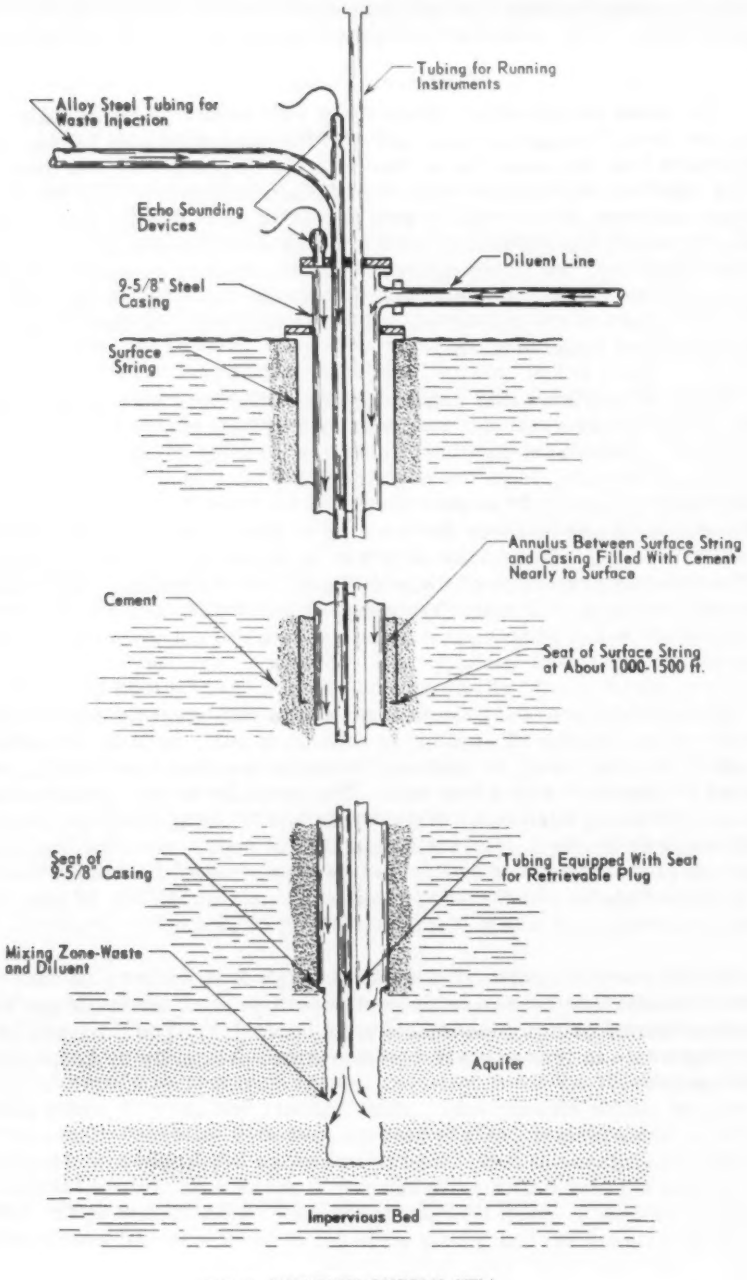
Tubing for Running Instruments
Alloy Steel Tubing for Waste Injection
Echo Sounding Devices
Diluent Line
9-5/8" Steel Casing
Surface String
Annulus Between Surface String and Casing Filled With Cement Nearly to Surface
Cement
Seat of Surface String at About 1000-1500 ft.
Seat of 9-5/8" Casing
Tubing Equipped With Seat for Retrievable Plug
Mixing Zone-Waste and Diluent
Aquifer
Impervious Bed

FIG. 1 – SUGGESTED DISPOSAL WELL

IX.

SITE SELECTION

The tremendous problem of transportation of liquid radioactive waste, as opposed to the relatively simple problem of transportation of spent fuel elements, indicates that the deep well disposal site should be near the fuel element reprocessing plant. As a practical matter, the location of the processing plant might well be determined by the availability of a site suitable for disposal.

In the selection of a deep well disposal site, a knowledge of the subsurface geology is essential. As discussed in preceding sections, the aquifer used for disposal must be of sufficient thickness and areal extent, and reasonably continuous with respect to permeability. Although porous limestones or fractured shales cannot be ruled out for disposal of waste, sandstones are preferred. Provided the permeability is reasonably high, the occurrence of some shale in the section is not objectionable. The thickness of the aquifer should be of the order of tens or even hundreds of feet. From practical considerations, it is likely that the area required for the disposal site, all of which must be subject to control by the authority responsible for disposal, should be in excess of 10,000 acres. It might even be desirable that the area be as much as 100,000 acres.

As discussed in a preceding section, suitable sand members may be lenses of limited area completely surrounded by impermeable formations, or they may be extensive beds, sometimes continuous to a surface outcrop, sometimes pinching out between impermeable strata without reaching the surface. Regardless of whether the sand chosen is of the lenticular type or is an extensive continuous member, adequate subsurface geological information must be developed, and this deserves much thought. In the preliminary stages of site selection, governmental agencies, including state geological organizations, industry, and professional groups can be of invaluable assistance. The best knowledge of subsurface geology is that which has been obtained by drilling deep wells for petroleum exploration and production. This knowledge, however, is more detailed in petroleum producing provinces, which, generally, are to be avoided in the search for radioactive waste disposal sites. Complete knowledge of all old wells drilled to the disposal horizon is necessary. It must be ascertained that such wells have been properly plugged and abandoned so that there is no chance that they might provide a channel for contamination of other horizons.

In the final stages of site selection, it will be necessary for the required subsurface information to be developed through a well planned

exploration program, for which the methods customarily used in exploring for petroleum may be used. These include studies of surface geology, geophysical exploration, and drilling of core holes. It should be noted that the core holes, properly cased and equipped, may be utilized later as monitoring and observation wells should the site prove suitable for waste disposal. Recovery of cores from the aquifer and the overlying strata permits the evaluation of the porosity, permeability, and mineralogic composition of the rock, and sampling of water native to the aquifer permits the determination of the compatibility of radioactive waste with this water. Testing the core holes also allows estimation of the capacity of the aquifer for water or waste injection and determination of the hydrostatic pressures at various points. Reliable pressure measurements at several points will reveal the occurrence of any natural fluid movements within the aquifer. The behavior of the wells on prolonged production of water from the core holes should be observed, to indicate the extent of the aquifer, and to predict how injected fluids may be expected to move. An important point which must be emphasized is that, even after a suitable site has been found, the necessary tests may require many months for completion.

While it is obvious that certain utilities such as electric power, water, etc., are required in the operation of a deep well disposal plant, there are other less obvious considerations in the selection of the site. If bleed wells are required to control pressures and pressure gradients in the disposal operation, then provision must be made for disposal of excess water from the bleed wells.

Even though there will undoubtedly be many problems connected with the selection of suitable sites for radioactive waste disposal, and even though the acquisition and exploration of a site will be expensive, it appears that, through a sound exploration program, suitable sites can be found.

X.

INJECTION PLANT

Although there is not sufficient information to design a plant for the disposal of radioactive waste, it might be desirable to describe a hypothetical plant in order to point out some of the design problems that will be encountered and to suggest some solutions. Broadly, such a plant might consist of two main systems:

1. A hot system for the storage, treatment, and filtration of high level waste prior to injection, and

2. A cold system for the producing, treating, and disposal of salt water for the purpose of conditioning the disposal aquifer to receive the waste and for providing suitable diluent for the waste solutions from the processing plant.

Fig. 2 is a flow sheet showing the major elements of such a hypothetical plant. The design of the first system would be governed by the technology of the chemical processing of radioactive materials, while the design of the second could be based on the technology and experience of the petroleum industry. In the disposal wells, these technologies overlap.

Obviously, much of the equipment and piping requires adequate radiation shielding. Some preliminary storage of high level waste is contemplated to permit the decay of some of the short-lived radioisotopes. This surface storage is expensive, costs quoted in the literature and reports ranging from about 17¢ to about $2.80 a gallon. Furthermore, there is a potential hazard associated with surface storage of such dangerous materials. One of the potential advantages of deep well disposal is that there is a chance that the storage period may be relatively short and the requirements for storage correspondingly small. In order to estimate the optimum storage time, it would be necessary to have data on the retention of radioactive ions by the disposal formation in order to estimate the maximum temperature to be expected in the neighborhood of the disposal wells.

The composition of the waste must be known in order to plan any chemical treatment that may be required. It will undoubtedly be necessary to provide for the removal of any suspended solid material from the waste, which would require the use of either filters or centrifuges. It may be that some of the filters used in the chemical and petroleum industries can be adapted to this service. The major problem in filtering the waste appears to be that of providing for backwashing of the filters and occasional replacement and disposal of the filter elements. In order to provide for flexibility in the operation of the plant, some storage of treated and filtered high level waste would be desirable.

The treated and filtered high level waste would probably have to be diluted considerably in order to avoid excessively high temperature in the neighborhood of the disposal well. As previously pointed out, there appears to be advantages in mixing the waste and diluent at the bottom of the disposal well, thus avoiding the need for facilities at the surface for handling large volumes of corrosive radioactive solution. The introduction of diluent serves the immediate purpose of keeping the disposal well and the surrounding cement cool. It also achieves the long-range objective of distributing the waste throughout such a large volume of rock that

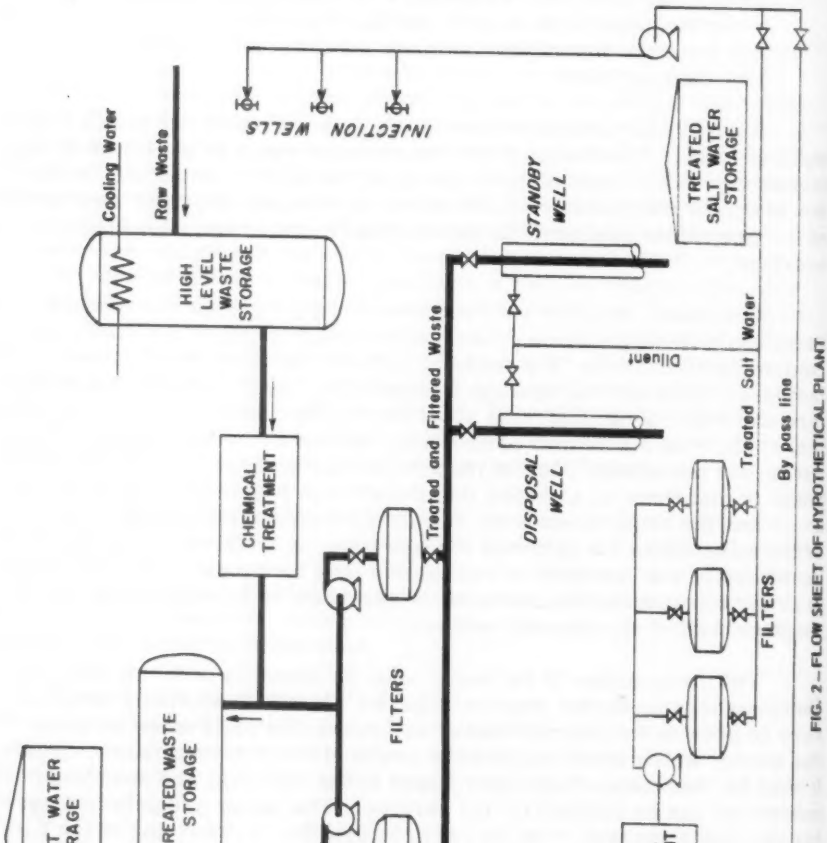

FIG. 2 – FLOW SHEET OF HYPOTHETICAL PLANT

the heat generated during the storage can be dissipated without developing excessive temperatures. The volume of diluent required to distribute the waste is much greater than that required to keep the well cool, but the distribution can be accomplished by injecting diluent at any time over a long period of years even after the disposal of radioactive material has ceased. For this reason, it may be more economical to design for moderate dilution, say 20 to 50 volumes of diluent per volume of waste, and to inject any additional diluent needed for temperature control in the aquifer only at such time and in such quantities as are found necessary by observation of the temperature developed in the rock. This might require that the disposal area be kept under observation after cessation of waste injection, and that standby water injection facilities be maintained until maximum temperature had developed.

Experience may show that there would be no danger in pumping some diluted waste directly into a suitable aquifer without any attempt to control the pressure in the aquifer or the manner of migration of the waste. However, the first installation will necessarily be in the nature of an experimental plant, and every precaution must be taken to insure safety of the operation and to provide for adequate control of the injected fluids. In order to do this, it is suggested that a number of auxiliary wells be incorporated in the disposal plant.

A suggested pattern of such wells is shown in Fig. 3. The most important of these are the bleed wells through which substantial quantities of salt water are produced. These are located roughly in a ring on the periphery of the area selected for disposal. An important function of these wells is to reduce the pressure in the disposal aquifer to such a degree that, if fluid migration of any kind takes place across its boundaries, it will result in a flow of cold salt water into the disposal zone, rather than flow of radioactive material out of the zone. The smaller the diameter of the ring of bleed wells, the more rapid is the pressure response within the disposal area and the better is the information on the distribution of the injected waste. On the other hand, the capacity of the system is less. The selection of the well spacing requires good engineering judgment to balance these factors but for the first installation a relatively small pattern is to be preferred.

If the bleed wells were located on the circumference of a circle of 2,000-ft. radius and if the aquifer had a net thickness of sand of 100 ft. and an average porosity of 20%, the fluid capacity of the disposal area would be roughly one and one-half billion gallons. Because of the vagaries of fluid flow through heterogeneous rock, all of this space cannot be effectively used for waste disposal, and the actual capacity of a system of such dimensions might be closer to one-half billion gallons of waste and diluent salt water.

If it be assumed that the plant is to handle 5,000 gallons per day of

FIG. 2 – FLOW SHEET OF HYPOTHETICAL PLANT

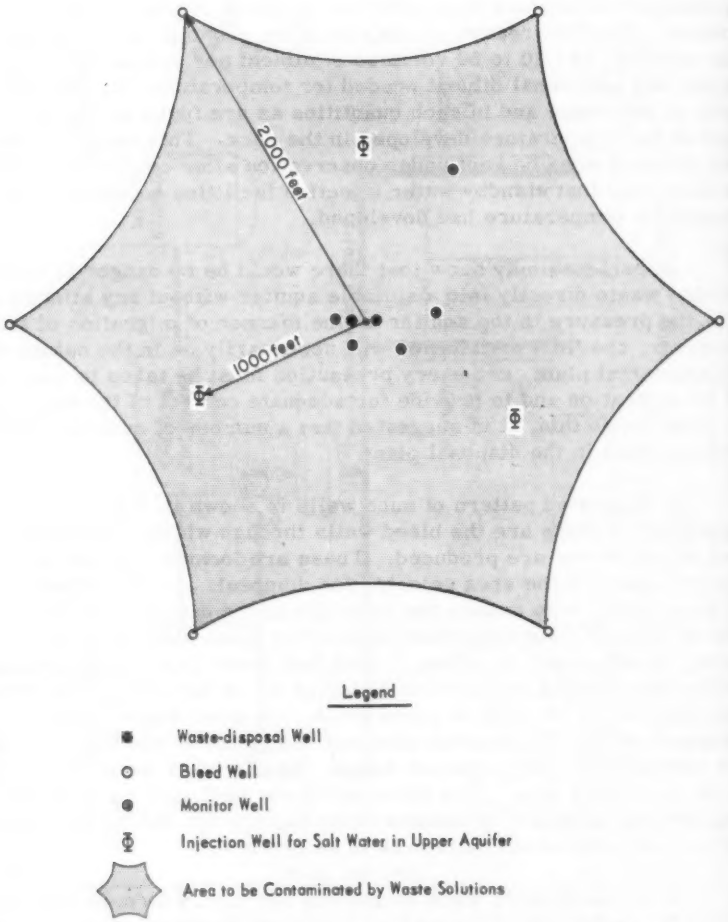

FIG. 3 – WELL PATTERN – HYPOTHETICAL WASTE-DISPOSAL PLANT

waste of such a level of radioactivity as to require dilution by a factor of 50, the disposal well would receive 250,000 gallons per day of fluids, and the capacity of the injection system would be reached in about five years. The disposal area could then be enlarged either by drilling more bleed wells further away from the disposal well or by developing an entirely new pattern of bleed and disposal wells. At the start of the injection, the area contaminated by the waste would be roughly circular, but will later develop into a scallop pattern with cusps extending toward the bleed wells. This pattern may be quite irregular and the cusps may not reach all bleed wells simultaneously, but the injected waste will not escape from the area of confinement without first appearing at one of the bleed wells. Furthermore, it should be noted that a high degree of control over the movement of waste can be exercised by adjusting the rate of flow of each bleed well. As soon as there is any indication of the approach of radioactive material, the bleed well affected would be shut-in pending enlargement of the capacity of the disposal system. There is some theoretical basis for expecting that the first waste products to appear will be the harmless hydrogen or nitrate ions, which would warn of the approach of more dangerous material.

It is emphasized that a major role of the bleed wells is to create a low pressure zone for the confinement of the waste. How this is accomplished is shown in Fig. 4, a plot of fluid pressure in the aquifer as a function of the distance from the injection well showing, under operating conditions, that the fluid pressure in the disposal zone could be lower than it was under the static original conditions. It would be advantageous, even though it may not ultimately prove to be economic, to reduce the pressure in the aquifer to such an extent that the disposal well would accept the desired quantity of waste by gravity flow, which would permit the surface equipment to operate under a vacuum. This might require a reduction in the pressure of the aquifer by several hundred lbs. per sq. in. and would require that water be produced from the bleed wells at a rate substantially in excess of the rate of injection into the disposal well.

The water not used as a diluent would have to be disposed of in some fashion. If the disposal site were near the coast, it might be possible to run a drainage ditch or other conduit to the sea. Alternatively, it would be necessary to dispose of the excess water by the more expensive method of injecting it into another aquifer, preferably one overlying the disposal site. A suggested arrangement of injection wells for water disposal is indicated also on Fig. 3. One important advantage of injecting excess water into an overlying aquifer is to increase the hydraulic gradient protecting the overlying formations from invasion by radioactive fluids.

In addition to the disposal wells, bleed wells, and water injection

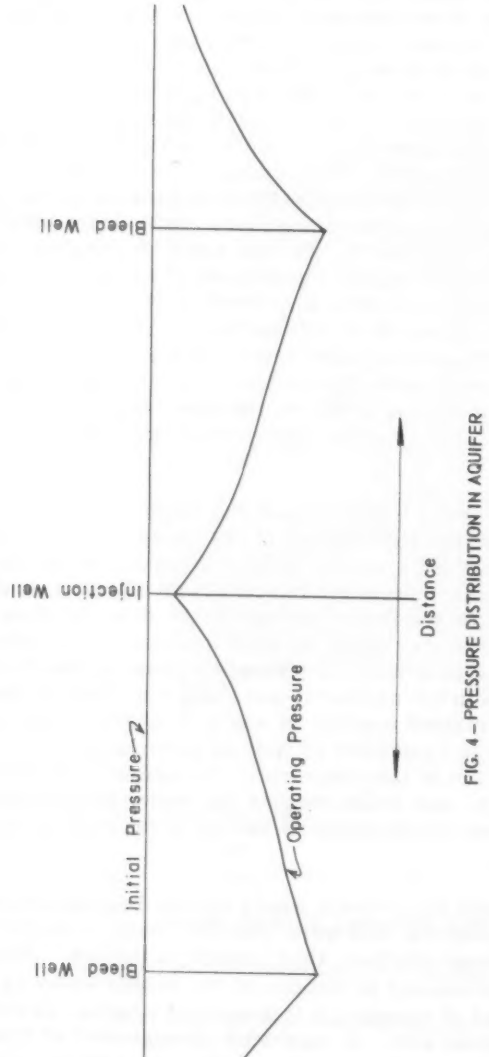

FIG. 4 – PRESSURE DISTRIBUTION IN AQUIFER

wells, observation or monitoring wells will be required in order to follow the spread of the waste disposal zone and to indicate what changes in the producing rate of the bleed wells might be necessary in order to produce a more desirable pattern of displacement. If these wells are to be used only for monitoring wells without any thought of converting them into disposal wells or injection wells later, they could be of small diameter and inexpensively equipped. They should be spaced at varying distances from the disposal wells and provisions should be made for measuring temperature, pressure, and radioactivity at frequent intervals.

The hypothetical plant described might well be more elaborate than is really necessary to put away the radioactive waste safely. However, in the first plant of this kind, it would be well to provide any reasonable safeguard, even though experience might show that the precautions were not needed and that a more economical plant could have been designed.

XI.

ECONOMIC FACTORS

It is well to restate the proposition that several methods for the disposal of radioactive waste might be developed, each of which might find application to certain types of waste and under specific geographic and economic circumstances. The selection of a particular process from several which are acceptable from considerations of safety will ultimately be made on the basis of economics.

It is obviously impossible to estimate accurately the costs of a disposal plant such as the hypothetical one described. Nevertheless, in arriving at a decision as to the merit of further development of the process, some rough feel for the cost would be helpful.

The order of magnitude of the investment in the disposal wells and the cold water system is indicated in the following table:

Bleed Wells, with High Capacity Submersible Pumps, 5,000' 6 at $85,000	$510,000
Disposal Wells, Stainless Tubing, 5,000', shielded 2 at $200,000	400,000
Injection Wells, 4,000' 3 at $50,000	150,000
Monitor Wells, 5,000' 5 at $40,000	200,000
Power Distribution System 2,000 Kw.	300,000
Cold Salt Water Lines, 4 mi. 6" d.	100,000
Storage Tanks, 100,000 bbls.	200,000
Pumps and Machinery, 1,000 H.P.	150,000
Cold Salt Water Filter & Treating Plant	100,000
Instrument and Controls	250,000
	$ 2,360,000

If it be assumed that the hot side of the system, i.e., the hot waste storage and filters, and any other facilities will require an investment of the same order of magnitude, the plant would cost in the neighborhood of five million dollars, exclusive of the cost of the site.

In making rough estimates of oil refinery costs, it is sometimes assumed that operating costs can be approximated by taking the sum of a fraction of the investment and a multiple of the operating labor. The operating labor in the plant might run about $135,000 per year. Guessing that operating costs, including depreciation, might be in the neighborhood of 22% of the plant investment plus 2.7 times the operating labor, the annual cost of operation is indicated to be about a million and a half dollars. This is equivalent to a cost of about 85¢ per gallon of high level waste, or about 2¢ per gallon of diluted waste.

XII.

SUGGESTED LABORATORY INVESTIGATIONS AND THEORETICAL STUDIES

Several of the problems anticipated in the deep well disposal of radioactive waste may profitably be investigated in the laboratory. Some techniques which may be useful have been developed and applied to the study of similar problems which are of interest to the petroleum industry. Among the more important problems are the following:

1. Ion Sorption

One of the problems amenable to laboratory study is that of ion fixation on the surface of the porous rock of the aquifer. Laboratory experiments conducted with long cores of naturally occurring sandstones should permit study of the amounts of adsorption of components of the radioactive waste which occur under various conditions of ionic concentration and temperature. Chromatographic separation of various ions present in the waste can be similarly investigated, and, if it appears desirable, attention could be given to application of the same techniques to the study of means for reducing adsorption by addition of more strongly adsorbed ions. Laboratory investigations of sufficient scope to define the problem and to furnish data of the required engineering accuracy might be completed by the expenditure of no more than about two or three man-years.

2. Plugging

Another problem which may be studied readily in the laboratory is that of the tendency for plugging of permeable sands as a result of interactions between injected waste and interstitial waters, and between injected waste and solid components of the aquifer rock. In such a study two types of experiments might be utilized; first, those in which possible

reactions leading to formation of precipitates may be investigated by very simple, conventional, bench-type methods, and second, those in which actual displacements of interstitial water by injected waste may be studied by flow through long cores of naturally occurring sandstones. If precipitates are found to be formed, then logically succeeding phases of the study might be concerned with the causes and the mechanism of the precipitation, with the effect of precipitation upon permeability of the rock, and, if so indicated, with means of minimizing precipitation and maintaining permeability. Particular parameters of interest are the effect of mineralogic composition of the rock of the aquifer, the compositions of interstitial water native to the aquifer, and the composition of the radioactive waste. The laboratory investigation of plugging would probably require two or three man-years to get sufficient data for preliminary planning of the disposal system.

3. Heat Dissipation

A problem which may be investigated theoretically is that of the rate of dissipation of heat generated in an aquifer by injection of radioactive waste, and the temperature to be expected in the aquifer. The petroleum industry, over a period of years, has developed a fund of knowledge in the formulation of the complicated mathematical relationships involved in similar problems and in their solution for particular geometrics by use of analog and digital computers. Values of thermal conductivities of earth materials have been measured experimentally, and data on heat diffusivity have been obtained in field tests of thermal processes for oil recovery. Use of these mathematical techniques, together with use of experimentally derived conductivity values, should permit the generation of a complete framework of the relationships of temperature rise in the aquifer to the geometry of the aquifer, to the specific activity of the injected waste, to the volume of waste injected, and to elapsed time.

The study of the problem of heat dissipation and temperature rise requires highly specialized skills on the part of the investigator and special computing facilities. For this reason, it might be desirable that the investigation be conducted by contract with experienced specialists. It is likely that one of the largest items of expense in the investigation would be that required for use of suitable computing facilities. In terms of over-all outlay, it is probable that the investigation would require $100,000 or more, with the manpower requirement dependent upon the extent to which computing machines are used effectively.

4. Corrosion and Radiation Damage

Still another problem which may be studied in the laboratory is that of corrosion and radiation damage to equipment and materials which

would be required in an injection facility designed for the handling of radioactive waste. These materials include those of which the well casing, tubing, and connections would be fabricated, and additional materials used in the construction of any packers, tools, and instruments required by the injection installation and chemicals used in the treatment of waste. Undoubtedly much more information on radiation damage has been compiled by the Atomic Energy Commission than is available from any other source, and the Commission also has at hand more extensive information on those construction materials which have been found suitable for use with radioactive waste than does the petroleum industry.

Nevertheless, the specific problems of damage to well equipment as a result of corrosion and radiation should be anticipated, and suitable protective measures devised.

XIII.

FIELD INVESTIGATION

In the event that a decision is reached to attempt to undertake a successful system for disposing of radioactive waste in deep wells, it will be necessary to pursue a program of field investigations to supplement studies in the laboratory. Such field investigations fall naturally into two broad categories: those concerned with the exploration for suitable disposal sites and those concerned with the development of a specific process. In this problem the design of the process depends on the site selected for disposal so that it would be advisable to carry out a limited exploration program concurrently with the process development.

In process development the chemical engineer has perfected the technique of using pilot plants to predict and test the performance of full scale plants. The equivalent of the pilot plant in waste injection operation is the shallow well which can be drilled cheaply and tested quickly. To the extent that it is possible to use such shallow wells effectively, substantial savings might be made in the cost of the development program. Unfortunately the dependence of the process upon the specific conditions at the disposal site is so great that some of the most important investigations of a pilot nature would have to be carried on at the specific site. Furthermore, it may be that the total cost of the pilot investigation is large in comparison to the cost of the wells, and if so, large savings might not be obtainable through testing in shallow wells. The selection of the scale and location for pilot testing will require keen engineering judgment to achieve a good balance between the value of the results and the cost of the test. Bearing these considerations in mind, some of the field investigations which might be considered will be discussed.

1. Survey of the Sedimentary Basins of the United States

It would be desirable to have a compilation of significant information

on all potential disposal reservoirs in the United States. Since these are most likely to be found in the sedimentary basins, it would be worthwhile to have a good description of the regional geology of each such basin in areas which might be considered for disposal operations. An effort should be made to describe the stratigraphic columns at key points in the basin. Isopach maps of the aquifers should be prepared, together with data on or estimates of the physical properties of the important rocks. One important part of such an investigation would be a consideration of the economic geology of the area and the nature of the surface.

In any study of this kind the most important information will be found in those areas which have been most thoroughly explored for petroleum or other minerals. While such areas cannot be definitely ruled out as possible disposal sites they would not be the most favorable areas from which to select the first major deep well disposal site. Thus the areas of greatest interest for disposal purposes are those where the information is scarce, but it would be desirable to assemble significant information on all sedimentary basins including that which may be available from private sources and which might have, at one time, been considered confidential. In making a study of this kind valuable advice and assistance might be obtained from professional societies such as the American Association of Petroleum Geologists. One area of particular interest because of its stratigraphy is the Atlantic Coastal Plain.

2. Preliminary Exploration

The development of deep well disposal will necessarily involve a program of exploration. The magnitude of such a program would depend, of course, upon the amount of interest in the method, either as a more economic means of waste disposal at existing processing plants or as a possible means of waste disposal at a processing plant planned for some specific immediate purpose. The exploration program envisaged should be quite flexible and would be carried on at a modest pace which would provide information on a specific site only when the information was needed to fit into the over-all pattern of the development. In this way the cost of the program could be kept low and it could be discontinued promptly in the event of selection of an unsuitable site or in case other investigations pointed out insurmountable difficulties in the deep well process. The exploration program might embrace some geophysical reconnaissance, such as a gravity survey or some air-borne magnetometer work over a limited area, selected on the basis of the regional studies and the requirements of the existing or projected processing plants with which the disposal operation would be associated. Some seismic work may be desirable. In many areas extensive geophysical data have already been obtained which might be made available to the Atomic Energy Commission. This preliminary geophysical reconnaissance could then be used in locating a suitable stratigraphic test well which would provide definite and specific information which would be essential to the proper planning of the process.

In drilling such a stratigraphic test the potential use of a particular aquifer as a disposal reservoir demands extreme care in completing or abandoning the well.

3. Test of Compatibility of Waste with Reservoir Rock and Fluid

One of the major problems associated with deep well disposal will be concerned with the problems of undesirable reactions of the waste solutions with the rocks and fluids in the reservoir. The kinds of problems to be expected and something of their magnitude can be learned from a laboratory study, but at some state of the development the compatibility of the waste and the reservoir should be tested in the field. If these problems appear to depend more upon the nature of the waste than on the nature of the reservoir rock and fluids and if it appears that they can be tested by the injection of a limited quantity of waste, the advisability of running tests in shallow wells should be considered. On the other hand, if the problem arises as a result of conditions in the disposal aquifer, the tests should be run in a well drilled especially for the purpose at the particular site being studied. Tests of compatibility of the reservoir can be run on cold simulated waste. Whether to use such waste or whether to use actual waste from a processing plant would be largely a problem of cost and the need for using actual waste in the investigation of other related problems.

4. Distribution of Radioactive Material

The dissipation of heat from the radioactive material is a problem which depends to a considerable degree upon the manner in which the radioactive material is distributed. This depends in part upon the way in which the radioactive ions are adsorbed or otherwise fixed in the rock at varying distances from the well. It depends in part on the way in which the injected solution displaces the salt water from the aquifer. The distribution of the radioactive material can be estimated from the results of tests involving the injection of radioactive waste in one well and sampling the fluids and running logs in a nearby well. These tests should be run with radioactive materials, but for preliminary work it would be desirable to use low-level solutions. Some information could be obtained by carrying out this work in shallow wells, but it may well be that the greater advantage of obtaining more complete information on the actual disposal reservoir would justify the increased cost of drilling the test well on the disposal site. One advantage of this is that there would be an opportunity for testing waste of successively higher levels of radioactivity, concluding with a full scale test of the disposal of the actual product to be handled.

5. Tests of Aquifer Prior to Operation

The final field program of experimental work would involve extensive testing of the disposal system and the disposal aquifer before the

injection of any significant quantity of high-level waste. This would require the drilling of bleed wells in addition to the disposal wells and the installation of the majority of the full scale disposal facilities. This phase of the work would be undertaken only in the event that favorable results had been obtained from all preliminary work and it would have as its object a final check on the safety of the disposal operation. It would involve careful pressure measurements in all available wells in the reservoir in order to detect any significant natural movement of ground water through the aquifer. It would involve the production of substantial amounts of water from the aquifer to learn something about its storage capacity and the rates at which waste could be injected without exceeding reasonably safe pressure limits. It would provide an opportunity for testing the formation and its confining beds to make sure that they would not be fractured at any operating pressure likely to be attained during the operation of the plant. This final phase of the testing program would be the most expensive part of the whole operation, and it is suggested here primarily to indicate how final tests of the feasibility and safety of the disposal system can be made.

XIV.

CONCLUSION

The subcommittee, on the basis of its consideration of the problem of disposing of radioactive waste in deep wells, has reached the following conclusions:

1. The disposal of radioactive waste from fuel reprocessing is a significant element in the cost of atomic power. While safety is the paramount consideration in the selection of waste disposal methods, cost is the factor that will govern the choice of safe disposal processes. The economic aspects of waste disposal will receive greater attention when atomic energy begins to compete economically in the market for industrial and domestic power.

2. Radioactive waste can be disposed of safely by injection, through deep wells, into porous rocks. There is, obviously, a limit to the amount and activity of waste that can be stored in any particular formation without danger. This limit cannot be accurately determined on the basis of available information, but the data required for estimating the safe storage capacity of a reservoir can be obtained experimentally.

3. Major problems in the disposal of radioactive waste through deep wells will be those of confining the waste, dissipating the heat generated, and protecting the disposal system

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against damage by corrosion or radiation. These will present difficulties, but no insurmountable obstacles are indicated.

4. A disposal plant might comprise, in addition to the injection wells, a number of auxiliary wells which serve various functions, with facilities for chemically treating, filtering, and storing waste and diluent water. Although reliable cost estimates cannot be made on the basis of information now available, preliminary consideration suggests that the cost of disposal might run less than a dollar per gallon of high-level undiluted waste.

5. A modest program of laboratory work and theoretical study would clarify many of the problems. If such a study confirms the preliminary conclusion that deep well disposal might be economic under favorable circumstances, a concurrent program of field testing and exploration should be the next phase of the development of the process to be undertaken.

Representative HOLIFIELD. We will ask Mr. Struxness to come to the stand again.

Some of these witnesses know so much we have to have them back two or three times.

Proceed please, Mr. Struxness.

STATEMENT OF E. G. STRUXNESS,[6] OAK RIDGE NATIONAL LABORATORY

Mr. STRUXNESS. Mr. Chairman and members of the committee, the feasibility of direct disposal of radioactive wastes to salt cavities and deep wells is being investigated at Oak Ridge. If these direct disposal methods prove to be safe and economical, it is important to consider the availability of suitable geologic formations in the selection of a site for any new power reactor fuel reprocessing plant. It is also desirable to study more than one method of direct disposal and to choose, at least for one plant, a site where both methods might be used. Then, with the experience which will come from actual disposal operations, each method can be given that part of the disposal burden for which it is best suited.

Dr. Heroy has explained why disposal in natural bodies of salt holds such promise, and has told you of the search he has made for locations suitable for field experimentation. The field experiments are needed to confirm the results of our laboratory work, and the work of others, showing that the chemical and physical reactions between present day waste types and rock salt will not significantly alter the structural integrity of a salt cavity containing radioactive wastes.

The chemical aspects of salt disposal are being investigated in the laboratory and, though the work is not yet finished, Purex and acid aluminum nitrate wastes appear to be essentially inert toward rock salt. It was found that these wastes dissolve some salt, but quickly become saturated. The solubility of salt is only slightly temperature dependent, so temperature changes are relatively unimportant in this respect. There is some chemical interaction betwen acid or neutralized waste and rock salt, resulting in the formation of small amounts of chlorine, nitrosyl chloride, and nitrogen dioxide gases, but not more than will remain in solution at moderate pressures. Radiation in the waste liberates hydrogen and oxygen, but the volumes do not appear to be troublesome. No other chemical effects have been observed and, though the waste salt mixtures are corrosive, rock salt appears to be able to contain them.

Some of the effects of radiation, heat, and pressure on the physical properties of rock salt have been studied on small test specimens. The effects of radiation are negligible at least up to exposures of 5×10^8r. The plastic flow of salt becomes significant at temperatures of 200° C. (392° F.), and above, and at pressures of 10 or more atmospheres.

For reasons of safety, it is desirable that the temperature of the salt surrounding the cavity be kept below 200° C. Hence, consideration must be given to the rate of heat production in the waste due to fission product decay and the rate of heat conduction in the salt surrounding the cavity. Calculations to determine the temperature distribution around a cavity are possible if certain assumptions are made

[6] See p. 1844.

as to the characteristics of the waste and the shape and size of the cavity. We have found that a spherical cavity 10 feet in diameter could contain 1 year old waste, having an initial heat production rate of 5 watts per gallon, without overheating.

Laboratory tests on the permeability of salt have not been conclusive. This was not surprising because we had been warned by geologists of the difficulties of making reliable laboratory determinations of permeability on so-called "undisturbed" rock or soil samples. Several salt mining companies provided us with large, specially mined blocks of rock salt. Some of these when tested appeared to be impermeable; others developed fractures, probably due to the release of the confining pressures of the earth. Determinations of the permeability of salt will have to be made on rock salt in place, either in mines or in boreholes.

Figure 1 is a scale drawing of a test cavity that will be excavated in the floor of a salt mine and the associated equipment with which we intend to conduct preliminary field tests with synthetic, nonradioactive wastes.

If you will refer to the diagram I will identify the components. All of this equipment and the cavities shown are in the floor of an existing mine. Above the cavity is a gastight cover. Off to the right are the various devices used to collect, condense, and permit the collection and analysis of off-gas constituents after which they are returned to the cavity.

Over on the left you see the heater controls and the temperature recorders.

Representative VAN ZANDT. Where is this mine located?

Mr. STRUXNESS. At present we are negotiating with the Carey Salt Co. of Hutchinson, Kans., for the use of their Hutchinson mine for this test.

The field tests will provide information regarding—

1. The structural integrity of waste filled cavities in salt;
2. The possible movement of radioactive fission products from the cavity out into the salt;
3. The migration of the cavity itself due to dissolution and recrystallization of salt which could result from the recirculation of vapor and liquid within the cavity;
4. Temperature gradients in the salt surrounding the cavity; and
5. Gas production under field conditions.

It is planned that these tests will begin in the next fiscal year.

I would like to state, Mr. Chairman, that our laboratory work and these preliminary field tests are designed to consider the worst possible case for salt disposal, namely, the storage of radioactive liquid wastes in the salt.

The problems are quite a bit simpler if we consider the storage of solid waste concentrates in salt.

Representative HOLIFIELD. Of course, your trouble of putting them in the solids is another problem.

Mr. STRUXNESS. Yes.

Dr. Moore has reported to you the view of the American Petroleum Institute committee on the disposal of radioactive waste into deep

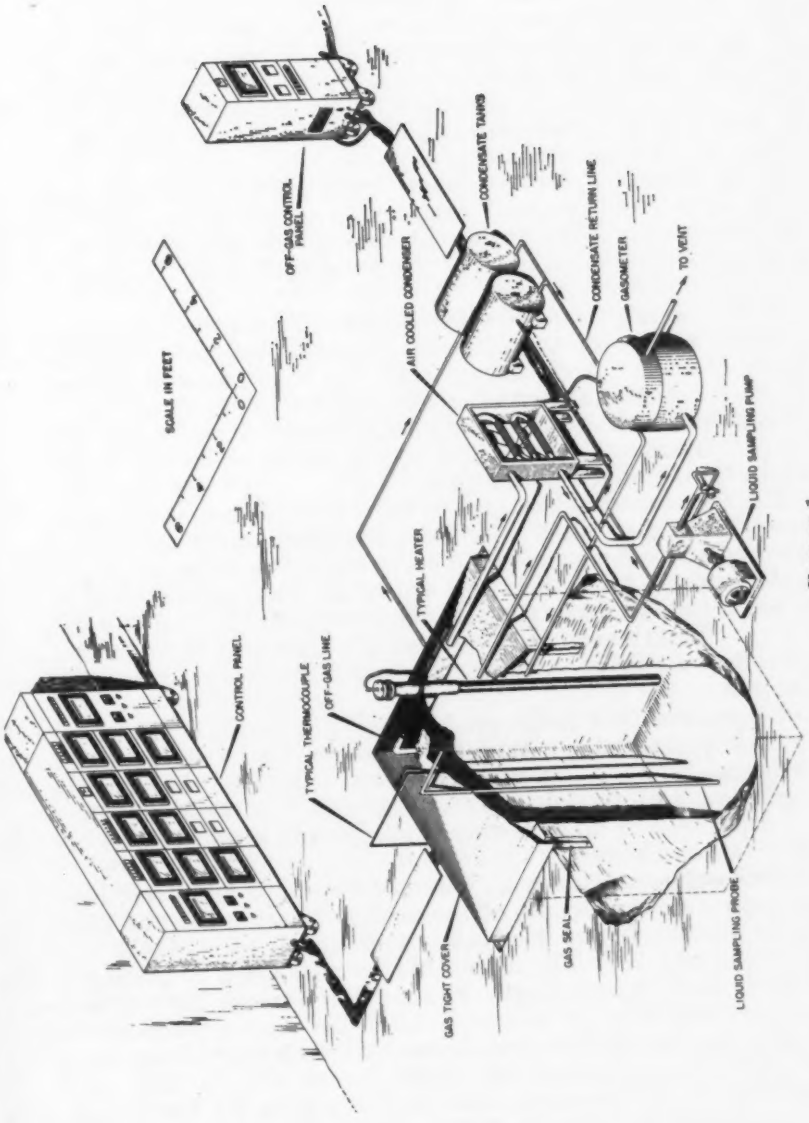
OFF-GAS CONTROL PANEL
CONDENSATE TANKS
CONDENSATE RETURN LINE
GASOMETER
TO VENT
SCALE IN FEET
AIR COOLED CONDENSER
LIQUID SAMPLING PUMP
TYPICAL HEATER
CONTROL PANEL
TYPICAL THERMOCOUPLE
OFF-GAS LINE
GAS SEAL
GAS TIGHT COVER
LIQUID SAMPLING PROBE

FIGURE 1

wells. We are in complete agreement with the committee's findings and wish only to add briefly to their report.

It is apparent that deep well disposal is most likely to be of use in handling large volumes of waste, where the large capital investment required for preliminary underground surveys, land, and the well field itself, can be justified. The method may be designed to handle high-level liquid waste, but it is more likely that it will be best suited for the larger volumes of medium to low-level waste that are also anticipated. Like disposal into salt, this method places severe requirements on the choice of location since the geology must be favorable. In this respect it may eventually supplement disposal into salt by widening the possible choice of localities.

In the laboratory we have learned how to treat simulated, high-level waste of several types with complexing agents such as citric and tartaric acids so that the waste will remain relatively free of suspended solids when neutralized. The same methods could be used to treat medium or low-level waste.

This is an important step forward, for the possible plugging of the disposal well or of the disposal formation has been regarded as a major obstacle. A significant start has been made in learning how to control the adsorption of cesium and strontium by the use of competing ions such as sodium and hydrogen. This control is needed in the immediate vicinity of the bottom of the well where intense heat production would be undesirable.

The thermal considerations in deep well disposal initiated by Professor Birch for the NAS–NRC Committee on Waste Disposal are being refined and expanded. We are computing temperature gradients in the geologic formations adjacent to the disposal aquifer in relation to fission product decay functions more representative of power reactor wastes. These calculations are being performed with an electronic computer which permits a more complete study of the variables involved.

The work at Oak Ridge on deep well disposal is developing slowly because the method is regarded as a possibility for the distant future. Furthermore, it is likely to continue at this pace for some time because development of this method will require the corporate knowledge and experience of many technical groups and governmental agencies, both State and Federal.

Representative HOLIFIELD. The fact that your deep well disposal is developing slowly because of the method in regard to the possibility for the distant future does not impress me at all. Maybe it should be developed a little bit faster. Maybe we should be getting a little bit more work done on that. You may need to ask for a little money to get on with this job because I think that these permanent stainless steel tanks and aboveground disposal methods that we are using now are pretty expensive and we might, by spending a little money on research and development, develop this a little bit faster and bring it into the near future in place of the distance future.

Representative VAN ZANDT. Has information been brought out at these hearings as to how much money we have invested in the present storage facilities?

Representative HOLIFIELD. We had the figure, I believe, of some $26 million in some of the early testimony.

Do you remember what the figure was?

Mr. STRUXNESS. No; I am sorry. I don't remember.

Representative HOLIFIELD. Mr. Lieberman.

Mr. LIEBERMAN. The total development in all waste-handling facilities is estimated at about $200 million at the present time, of which, as I recall, there is something like $115 million in waste storage facilities. This is the piping and so forth that goes with it, the tanks and so forth, about $35 million in the gas-handling facilities, and the remainder in the low-level waste and the waste handling. These are approximate numbers.

Representative HOLIFIELD. We are really in this business.

Representative VAN ZANDT. We are.

What percentage of the $200 million is represented by the facilities at Hanford?

Mr. LIEBERMAN. As I recall, and you can check me on this, the total investment in Hanford is $114 million, and I think they have about something close to $75 million in their tank storage facilities.

Chairman DURHAM. They still have some vacant space, of course, where they can store more?

Mr. LIEBERMAN. Yes, sir.

Representative PRICE. I would like to have some comment from Mr. Struxness on your apprehension about the slowness of the development of the program. What is the reason for this approach?

Mr. STRUXNESS. Part of what I was considering in making that statement was this: We at Oak Ridge National Laboratory will have very litle, if anything, to say about the choice of location. This will be done by geologists and geophysicists, people who are familiar with this type of environment. It will be done, I am sure, in close coordination with the U.S. Geological Survey. To some extent I suppose the State geological surveys will be involved, and so on.

What I am a little bit apprehensive about is coordinating this corporate effort and actually getting it started, getting the preliminary explorations started. We cannot take any chance on injecting wastes even in a test well until we know completely the mechanics of this reservoir. That was what I had in mind when I made this statement.

Chairman DURHAM. How much money did you spend on your project last year, do you recall?

Mr. STRUXNESS. Seventy thousand dollars.

Chairman DURHAM. Do you have anything in your budget this year for your project, that you know of?

Mr. STRUXNESS. This consists mainly of laboratory work by chemists who are studying the interaction between the wastes and the brines and the solid materials that we expect to encounter down in these formations.

We will continue this work for some time. We can finish perhaps all of it, certainly a large part of it, before any location will have been selected and the reservoir explored.

Chairman DURHAM. How do you operate? Are you in a position to go out and hire your geologists and your geophysicists to determine this strata in a given locality in which you have some thought about placing one of these places, or does that have to come from some other source?

Mr. STRUXNESS. I believe the Commission, working closely with the U.S. Geological Survey and the State geological surveys, would enter into the decision as to where this test would be run and would actually do the exploratory work.

I do not believe that our Laboratory would have very much to do with the preliminary explorations.

Chairman DURHAM. You have to have that determination made first?

Mr. STRUXNESS. That is correct.

Where we would expect to come into the picture later on is when we actually run a test.

Chairman DURHAM. You are running it at the tail end of it and the other people, the people who are making the determination on radiation, go ahead of you. You are at the bottom of the totem pole.

Mr. STRUXNESS. We could pursue it with more vigor but I doubt that it is necessary to expand our efforts too much at this time. I think that decisions have to be made elsewhere on where we might run some of these tests.

Chairman DURHAM. There is no use going out and spending $100,000 in a place when you don't know whether the geophysicists or the geologists will say that it is safe to put it there.

Mr. STRUXNESS. As you said yesterday, Mr. Durham, we better know what we are doing before we do it.

Representative HOLIFIELD. I think this points my thought that there has to be coordination of this by the AEC to bring these different studies together and, if necessary, accelerate some of them in order that we might in the first place start locating any new processing plants that are going to be necessarily close to a proper place of disposal, and also make arrangements to take care of the fission material that we will have in the interim.

Mr. STRUXNESS. Yes.

Chairman DURHAM. I agree with Mr. Holifield. This thing is growing on us day by day and night by night also. I think we have to get along with the job and make up our minds.

Representative HOLIFIELD. We have a number of these reactors going on the line in 1961, 1962, and 1963.

Representative VAN ZANDT. Fifteen of them?

Representative HOLIFIELD. Seventeen of them that are now being operated or constructed at this time and, of course, you have this Euratom program starting, and we will have some of the waste limits from that coming back to the United States for processing and burial of the waste material.

Even if you went into this thing on a crash program basis, you would just about be ready to start handling it by the time you are going to need to start handling it.

Mr. STRUXNESS. If you are interested, I might give you this figure. This year we are spending $350,000 on our salt investigations in contrast to the $70,000 we are spending on deep-well disposal.

Chairman DURHAM. Are you spending that now on the basis of the geologists and the determination being made that these places are going to be worth anything to you?

Mr. STRUXNESS. No, sir. The problem in salt is much easier. All we have to do is find the suitable facility already mined out and

conduct these field experiments, so we are prepared to begin these very soon.

Actual construction of our first test cavity will start within a month. The first test will start by July.

Chairman DURHAM. I don't know how much salt there is in the country, but certainly we don't want to destroy all the salt now because that is quite important.

Representative PRICE. These will be in abandoned mines, will they not?

Mr. STRUXNESS. The first test will be run actually in an active mine.

Representative PRICE. In an active mine?

Mr. STRUXNESS. Yes.

Chairman DURHAM. I don't understand that. Why is that?

Does he get more out of selling salt that way than he will by putting salt up in boxes?

Mr. STRUXNESS. I might say that these first tests will not involve radioactivity and we locate the test cavity away from the active workings of the mine so that we do not interfere with salt production.

Representative PRICE. How are you going to make an adequate test?

Mr. STRUXNESS. In the preliminary test, we are interested primarily in the heat distribution, the heat conduction away from the cavity, and also the possible migration of the liquid out into the salt. So we will have some 120 thermocouples and moisture indicators in the salt surrounding the test cavity.

Representative PRICE. I thought one of the important things also was radiation. Where are you going to get your radiation?

Mr. STRUXNESS. Later on we plan to use tracers in several cavities. But many of the answers in the migration of fission products through salt we think we can find out in the Laboratory working with small test specimens.

Representative VAN ZANDT. Have you looked at the salt mines in northern Pennsylvania?

Mr. STRUXNESS. Dr. Heroy, employed as a consultant to our Laboratory, looked at most all of the active salt mines in this country. We were quite surprised to learn that the International Salt Co. and the Carey Salt Co. were willing to let us use their property to run these preliminary tests.

Representative PRICE. Incidentally, where are the major number of salt mines in the country?

Mr. STRUXNESS. I am sorry, Mr. Price. I am not an expert in that field. I would rather if Mr. Heroy would answer that question.

Representative HOLIFIELD. Mr. Heroy.

Mr. HEROY. There are two in New York State: one near Ithaca and one near Canandaigua, which is south of Rochester. The one south of Rochester is much the larger and I presume probably the largest salt mine in the United States in the point of production.

There is one in Michigan, in Detroit or under Detroit. There are three in Kansas that are operating at the present time, the north and south mines extending from somewhat west of Wichita across the State.

There are two that are operating in Texas and then there are, I think, four in Louisiana that are also operating in salt domes. There are no abandoned salt mines in the gulf coast.

Does that answer your question?

Representative HOLIFIELD. Yes; it does.

Representative PRICE. I can't challenge it, that's a cinch.

Mr. STRUXNESS. Getting back to the level of effort at our institution; and, by the way, work is also going on at the University of Texas on disposal in salt, as we go from laboratory studies with synthetic studies to laboratory studies with actual wastes and to field tests, Congress can very well expect requests for more money.

Chairman DURHAM. Are they under direct contract at the University of Texas with the AEC?

Mr. STRUXNESS. Yes, sir.

Chairman DURHAM. That is the only one you have outside of your own work?

Mr. STRUXNESS. I believe that is correct, sir.

Representative VAN ZANDT. Are you familiar today with what we have in the way of available storage facilities to meet the future demand and how many years does that future represent?

Mr. STRUXNESS. I am told, and I get this figure from Mr. Heroy again, that in the present day enough space is mined out of salt to take care of all of the high-level wastes we would expect to produce between this year and the year 2000.

Representative VAN ZANDT. This is storage in the salt mine?

Mr. STRUXNESS. Yes.

Representative VAN ZANDT. What about the existing storage facilities?

Mr. STRUXNESS. The tank storage?

Representative VAN ZANDT. Yes. I think the chairman mentioned a moment ago that we have a surplus at the present time, due to the facilities are not filled up.

Mr. STRUXNESS. Essentially 90 or more percent of the liquid wastes in this country are at Hanford, and there are Hanford people in the room. Perhaps one of them could answer that question.

Mr. LIEBERMAN. I think I can give a part of the answer.

The total quantity of high-level waste in storage has been estimated at somewhere between 60 and 65 million gallons. As I recall, the total available tankage in this country is something over a hundred million gallons; I think 103 million or 105 million.

Representative VAN ZANDT. Then we have about 65 percent of our present storage facilities filled?

Mr. LIEBERMAN. That is about right.

Representative VAN ZANDT. With waste material?

Mr. LIEBERMAN. Yes, sir.

Representative HOSMER. Does it not seem that whatever method is used we probably should be looking toward using it somewhere near the Hanford area where most of the waste is accumulated to avoid the transportation problem, or should we look to the future as to where we are going to have the most atomic energy generated?

Mr. STRUXNESS. The committee will hear later testimony on the interest that many people have of the possible utilization of fission products in these wastes.

This is important to consider. Until we get some idea of the potential utilization of these fission products and until we have developed

completely these ultimate methods of disposal, I strongly suspect that we will continue to store these wastes in tanks.

Representative HOSMER. You think it is a good idea then because it is readily available if we do want to get the fission product out for some usable purpose?

Mr. STRUXNESS. I personally believe that we will be up to our necks in fission products some day and we will have to develop these disposal methods, but certainly one should consider the potential value of these things as well.

It is also true, I am told by the people who are interested in fuel processing and who know the tank-storage problem better than I, that tank storage is economic, that at this time it is possibly the cheapest method we have of storing these wastes.

Chairman DURHAM. We recover about what? Four or five percent of it, and store the 95 percent of it in tanks.

Mr. STRUXNESS. It is true, the market today is largely unexplored, but there are private companies that are looking at the potential market for these fission products and I believe you will hear testimony later today on this very subject.

Representative VAN ZANDT. May I ask Mr. Lieberman another question?

Representative HOLIFIELD. Surely.

Representative VAN ZANDT. With respect to this 40 percent, we will say, of excess storage space available at the moment, how many years will it be before such space is filled?

Mr. LIEBERMAN. I don't know whether I can answer your question directly. I will try. I think one of the things we have to remember is the tank space that is presently filled as a result of the past production program for plutonium, and one has to keep in mind that at least in the immediate future the waste that will be produced in the power program will probably be at a much smaller rate than that which is associated with the production program.

At the moment I am not prepared to assign specific numbers to this. On the other hand, I think we can say that we have accumulated the 65 million gallons over the life of the atomic energy industry as we know it.

This is perhaps 14, 15 years at Hanford, and 6 or 8 years at Savannah River, and roughly the same at Idaho, so if you wanted to make a straightline extrapolation, the 40 percent would be filled in 40 percent of the time that is already elapsed.

Representative VAN ZANDT. Would you say 10 years is a good guess?

Mr. LIEBERMAN. No, sir; I don't think I would because the point has been made a number of times already that in the whole chemical reprocessing business—it might be that sometime in the future, but I don't think this is in the immediate future—we might be able to find out a better way to process these fuels and we will not have such large volumes of these wastes.

However, I think it is probably fair to say at least in the next 10 years processes that are now being used will continue to be utilized, but at the same time there is a substantial effort, and I think it is meeting with considerable success, to reduce the unit volume of waste that is produced, so that if I were to say that in 10 years this 40 percent would be filled up, I don't think I would be quite correct.

I think it will probably be a longer time than that.

Representative VAN ZANDT. The purpose of the question was to then ask you, Mr. Struxness, in view of the fact that we have, we will say, 10 years ahead of us as far as space is concerned, do you think our present rate of effort in research and development is adequate?

Mr. STRUXNESS. The development of disposal of liquid waste in salt, I personally believe, can be worked out in the next 2 or 3 years. It is not so much whether it can be done. It is just how to do it.

The deep-well disposal I would hesitate to guess. I suspect that once a site is selected it might require the geologists and the geophysicists and the petroleum engineers and so on, 3 years of exploratory work before they would even let us conduct a test.

This would mean then, that if we pursued vigorously the selection of a site for deep-well experiment, we might be ready to run a test in 3 or 5 years and have some significant results in 10.

In the case of salt, I honestly believe that we could work this out in the next 3 years.

Representative VAN ZANDT. That is all.

Representative HOLIFIELD. I might, for the record, say that last August 4, we passed a bill providing for $15 million on the project 59–A–1 for plant modifications and for processing of nonproduction spent fuels at undetermined sites.

I don't know how that has been spent or whether the sites have been selected, but certainly, some of these factors ought to go into selecting sites.

At ARCO we built a new waste storage installation costing $3,200,000. There is a total of $18 million that we put in last year's authorization bill, and we have not seen what they are going to ask for this year.

It shows that we are dealing with some pretty substantial amounts.

Are there any further questions to Dr. Struxness?

If not, sir, we thank you.

Again we have additional papers for the record, and without objection, I will insert them at this point.

(The material referred to follows:)

STATUS REPORT ON WASTE DISPOSAL IN NATURAL SALT FORMATIONS

By

F. L. Parker, L. Hemphill, and Julian Crowell,+
Oak Ridge National Laboratory

The hazards created by radioactive fallout from the weapon-testing programs prompts an investigation into the source, magnitude, and disposition of the other radioactive debris generated in our nuclear age. The major source of radioactive wastes from peacetime uses of nuclear energy will be those produced by the irradiation of fissionable fuel in nuclear power reactors. The present rate of world-wide bomb testing has been estimated to be 10 megatons of fission bombs per year.(1) This rate could be continued with safety for the next thirty years, but should be reduced to 5 megatons per year for indefinite testing.(1) If the growth of nuclear power in the United States proceeds as predicted by Lane, we may expect to have 700,000 megawatts (thermal) of installed capacity by the year 2000.(2) Assuming a utilization factor of 100 per cent, we shall produce over five hundred times as much radioactive waste per year as we are producing in bomb tests. If we assume that world-wide usage of nuclear energy will approximate that of the United States by the year 2000, peacetime uses will generate a thousand times more fission products than bomb tests. The total peacetime fission-product load is actually greater, because megaton bombs send over half of their fission products into the stratosphere

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where the average airborne time is ten years, allowing for decay of many of the short and intermediate-lived fission products before their return to the earth by fallout.(3) Therefore, it is imperative that safe means be found to handle the radioactive waste products from nuclear reactors.

Sources of Radioactive Wastes

The major source of radioactive wastes in the future will be from stationary central power stations. Radioactive wastes from marine, research, test, irradiation, and package power reactors will be negligible in comparison. The fission products are formed when a neutron splits the fissionable atom into two unequal fragments (fission products) with the simultaneous release of neutrons and energy. Fission products, the principal radioactive constituents of reactor wastes, are produced in individual yields ranging from 0.00001 to 6.5 per cent. This is a function of the fissionable material and the energy of the neutrons. The fission products remain inside the fuel elements in a heterogeneous type reactor until the fuel elements are dissolved. If the nuclear economy were such that the fuel elements could be stored until the fission products decay, then the waste-disposal problem would be relatively trivial, involving only the safe storage of solid fuel elements. However, the present low burnup of the fissionable material, approximately 2-20 per cent, makes it necessary to process the fuel elements to recover the unspent fissionable material. If, in the future, high burnup rates can be achieved, chemical reprocessing of the fuel elements may not be necessary. It is also possible that the supply of fissionable material, at reasonable cost, may become so great that the reprocessing and recovery costs are greater than the cost of initial mining and processing. For the present, at least, it is necessary to reprocess the fuel elements to recover the fissionable material. Many

schemes have been developed to reprocess present-day fuel elements as well as proposed elements. The fuel reprocessing field is in a constant state of flux. Therefore, we have limited our interest, initially, to the two existing processes producing the largest volumes of waste. Eventually the other waste streams will be studied.

Waste Processes

The two processes of immediate interest are called Redox and Purex. Their characteristics, as well as those of other processes now in use, are given in Table I.[4] The fuel elements are removed from the reactor and cooled, allowing the shorter half-lived fission products to decay. The fuel element is then dissolved. The Purex and Redox systems, being aluminum clad and aluminum alloyed, use nitric acid. Stainless steel and zirconium fuel elements would have to be dissolved in aqua regia and gaseous HCl respectively. The fissionable and fertile materials are then separated from the fission products and cladding material by solvent extraction. The uranium is soluble in the organic solution and is separated from the aqueous phase containing the fission products and cladding material. This is repeated a number of times to gain the required purity for the reclaimed fuel. The Purex and Redox systems differ in their organic solvents and the salting agents used to help the separation, 30 per cent TBP in hydrocarbon and HNO_3, and hexone and $Al(NO_3)_3$ respectively. Over 99.9 per cent of the fission products are removed in the first solvent extraction. Table II[5] shows the chemical constituents of the first cycle extraction as well as the volume and activity of the waste to be expected.

Magnitude of Waste Problem

Based on the predicted growth of the electrical industry and the

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TABLE I. PROCESSES FOR CHEMICAL SEPARATION OF FISSIONABLE MATERIALS

Process	Organic Solvent	Aqueous Solvent	Irradiated Metal Feed	Salting Agent
For Separation and Decontamination of U and Pu				
Redox	Hexone	HNO_3	Natural U, Al can	$Al(NO_3)_3$
Metal Recovery	12.5% TBP in hydrocarbon	HNO_3	Caustic precipitated U, fission products	HNO_3
Purex	30% TBP in hydrocarbon	HNO_3	Natural U, Al can	HNO_3
TTA chelation	0.25 M TTA in hexone	HNO_3	Natural U, Al can	$Al(NO_3)_3$
For Separation and Decontamination of Enriched U and Al				
25	Hexone	HNO_3, $Hg(NO_3)_2$ catalyst	U-Al alloy	$Al(NO_3)_3$
25, TBP	5% TBP in hydrocarbon	HNO_3, $Hg(NO_3)_3$ catalyst	U-Al alloy	HNO_3-$Al(NO_3)_3$

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TABLE II. CHARACTERIZATION OF FIRST CYCLE HIGH LEVEL AQUEOUS WASTES FROM SELECTED SOLVENT EXTRACTION PROCESSES[1]

Process	Chemical Properties, Constituents (M), Approximate, Exclusive of Fission Products and Heavy Elements																	Waste Volume, gal/g U^{235} Consumed (4)	Approximate Specific Gravity	Approximate Concentration of U in Feed g/liter	Waste Activity[2 x 5]		
	H	Al	Fe	Cr	Ni	Zr	Na	NH_4	NH_3	Sn	Mn	Mg	Hg	NO_3	F	SO_4	PO_4	Cl				curies/gal	Total watts/gal[3]	BTU/hr/gal
Redox	~0.2	1.2	*	*	*		0.46							4.1					0.27	1.18	450	1720	8.6	29.4
Purex	2.5 to 7.0		*	*	*		*							2.5 to 7.0		*			0.34	1.07	300	1320	6.7	22.7
Hexone-"25"	~0.2	1.5	*	*	*			0.5					0.01	4.5					0.31 to 0.13	1.23	2-5	1620 to 3940	8.1 to 20.2	27.7 to 69.0
TBP-"25"	0.5	1.6	*	*	*								0.01	5.5		0.02	*		0.22 to 0.11	1.29	3-6	2580 to 5160	12.9 to 26.0	44.1 to 88.6
Zirconium-HF for enriched U	0.8 to 2.0	0.8		0.1		0.5				0.02				2.3	3.0				~1.50	~1.1	0.3	350	1.75	6.0
Stainless Steel-H_2SO_4 for enriched U-235	2.6	0.25	0.05	0.01	0.03		*				0.001			2.4		0.30			~0.35	~1.3	2-3	1500	7.5	25.0

Notes: (1) Wastes are untreated; they are essentially as they leave the solvent extraction plant and are subject to further treatment such as evaporation, neutralization, chemical treatment for fission product removal, etc.

(2) Basis for activity numbers: Irradiation period 4000 mwd/t for natural uranium
5×10^{13} $n/cm^2/sec$
4000 grams U^{233} chain per ton of thorium
53% burn-up for U^{235} in enriched fuel elements
100 days decay cooling from time of reactor discharge

(3) After 100 days' decay, the distribution of energy is approximately 50% γ and 50% β.

(4) Waste volume per gram U^{235} consumed is an inverse function of burnup; i.e., for hexone-25 at 20% burn-up, the gal/g U^{235} = (53/20) (0.14).

(5) Waste activity varies approximately as the (irradiation level)$^{0.2}$; and indirectly with the concentration of fissionable or fertile material in the fuel.

percentage of that which will be nuclear power, Zeitlin, et al. have predicted the waste volumes and activities to be expected if present processing schemes are used, Figures 1 and 2.(6) Assuming a waste volume of 820 gallons per ton of spent fuel reprocessed and a nuclear generating capacity of 700,000 thermal megawatts and a burnup of 4000 megawatt days per ton, there will be an accumulated volume of one-half billion gallons of waste by the year 2000 and 3.5×10^{11} curies of activity.

If the present system of storing wastes in tanks is continued, the operating and capital cost of storage at $1.00 to $3.00 per gallon for 20 years of waste storage quickly becomes astronomical.(7) The hazard associated with such large amounts of liquid wastes at the surface where they may be subject to earthquake, sabotage, and explosion is great. Therefore, cheaper and safer methods of storage of radioactive wastes are needed. This is a preliminary report describing the advantages and problems of one suggested method - ultimate storage or disposal in natural salt.

Disposal of Wastes on Land

At the request of the Reactor Development Division of the AEC, the National Research Council (Earth Sciences Division) called a meeting at Princeton University in September 1955 to discuss the disposal of high level radioactive waste products in geologic structures. The conference was divided into two groups, those concerned with disposal at depths less than 1000 feet below the surface and those concerned with disposal at depths of 10,000 to 15,000 feet. As a result of these deliberations, the order of feasibility of radioactive waste disposal in geologic structures was determined to be: (1) storage in salt beds, salt domes, abandoned salt mines, and related structures; (2) storage of ceramic material or a slag formed from high-level wastes in sheds on the

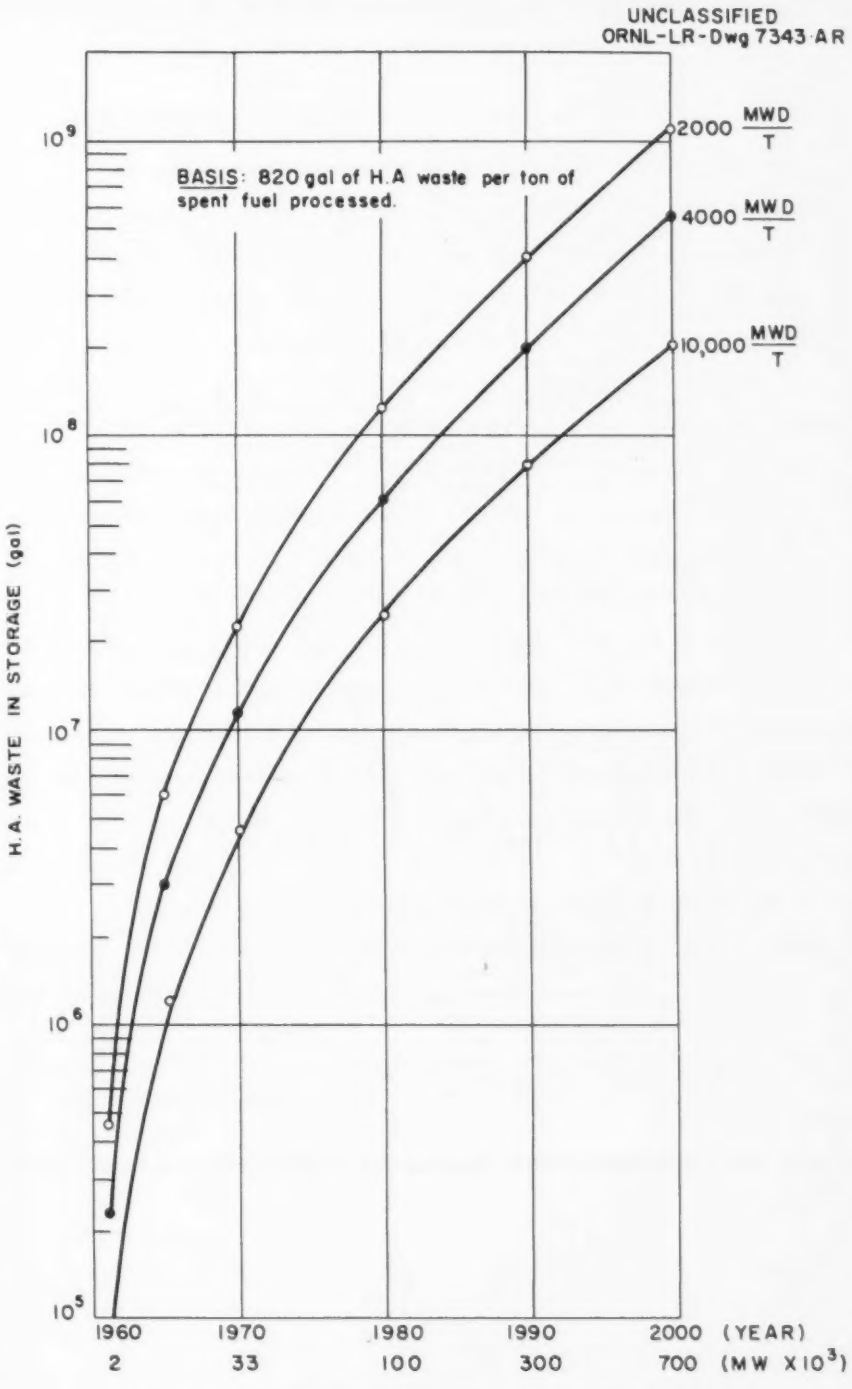

Fig. 1. Accumulated Volume of High Activity Waste.

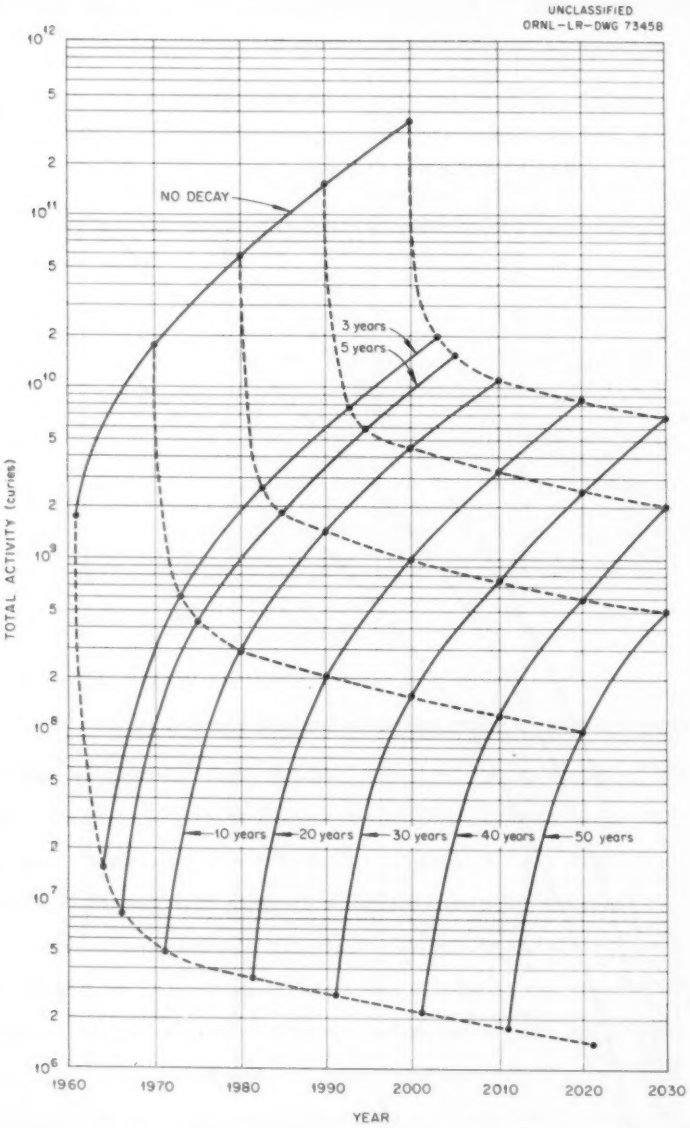

Fig. 2. Total Accumulated Activity Resulting from a Predicted Nuclear Power Economy.

surface in arid areas, or in dry mines; (3) disposal of the liquid wastes in porous media such as sandstone at comparatively great depths.(8)

Disposal of Wastes in Salt Cavities

The suggestion for the use of cavities in salt formations for the storage of radioactive wastes was prompted by the knowledge that the petroleum industry has been storing liquified petroleum gases economically and safely in salt formations since 1950. The first storage in dissolved cavities in salt beds took place at Hutchinson, Kansas, and Upton County, Texas, in 1949. However, it was not until 1950 that storage in the Keystone Oil Field of Winkler County, Texas, proved so successful that the use of underground storage in salt for petroleum products was widely accepted.(9) The growth in storage capacity since that time has been spectacular, from zero storage in 1950 to 7,000,000 barrels* in 1952, and over 25,000,000 barrels in early 1957.(8)(10)

The use of salt cavities for the storage of liquified petroleum gases was based upon lower costs and greater safety. Figure 3 shows comparative costs for surface and underground storage.(11) For large volumes dissolved or mined caverns are much cheaper per unit volume than above ground storage. The greater safety of underground storage from a strategic point of view is obvious. It provides the maximum possible protection against the hazards of our nuclear age.(12)

If the disposal of radioactive wastes in salt should prove equally successful, then one of the most vexing problems in the nuclear power complex would be solved.

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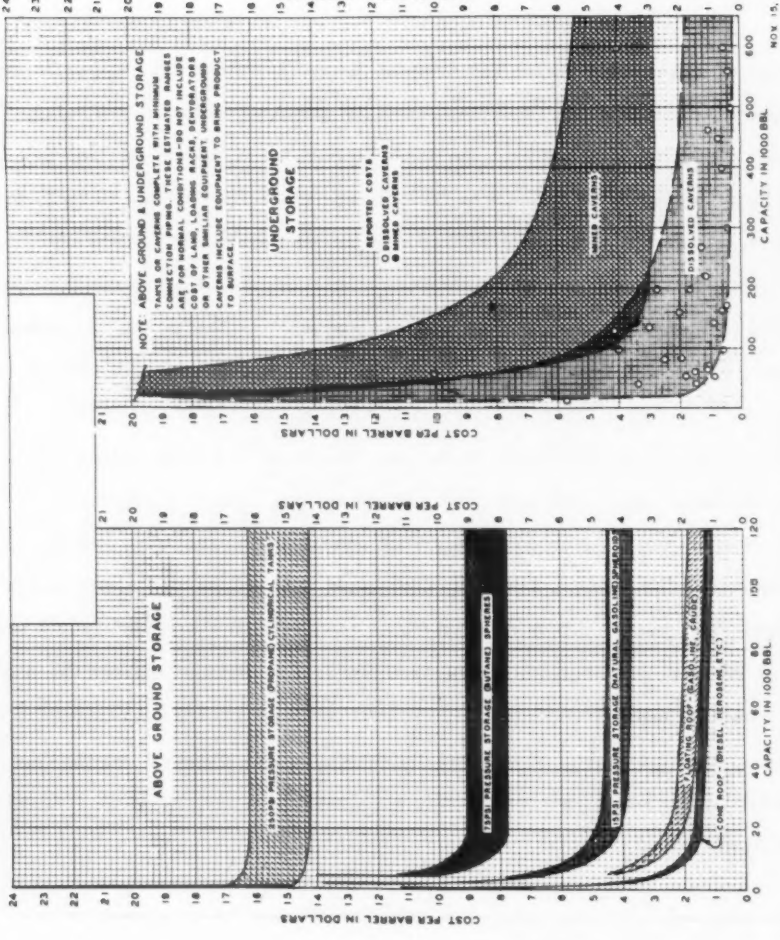
*42 gallon barrels.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ABOVE GROUND STORAGE
1525 PRESSURE STORAGE (BUTANE) SPHERES
1525 PRESSURE STORAGE (PROPANE) SPHERES
1525 PRESSURE STORAGE (NATURAL GASOLINE) SPHERES
CONE ROOF (DIESEL, KEROSENE ETC.)
FLOATING ROOF (GASOLINE GRADES)
CONE ROOF (GASOLINE GRADES)
CAPACITY IN 1000 BBL.
COST PER BARREL IN DOLLARS
NOTE: ABOVE GROUND & UNDERGROUND STORAGE
TANKS OR CAVERNS COMPLETE WITH MINIMUM CONNECTION PIPING. THESE ESTIMATED RANGES ARE FOR NORMAL CONDITIONS - DO NOT INCLUDE COST OF LAND, LOADING RACKS, GENERATORS OR OTHER SIMILAR EQUIPMENT UNDERGROUND CAVERNS INCLUDE EQUIPMENT TO BRING PRODUCT TO SURFACE.
UNDERGROUND STORAGE
REPORTED COSTS
DISSOLVED CAVERNS
MINED CAVERNS
MINED CAVERNS
DISSOLVED CAVERNS
CAPACITY IN 1000 BBL.
COST PER BARREL IN DOLLARS
NOV. 15, 1956

Fig. 3. Comparison of Construction Cost Ranges for Various Types of Product Storage.

Fig. 3. Comparison of Construction Cost Ranges for Various Types of Product Storage.

Advantages of Disposal in Salt Cavities

The proposed use of salt formations as a storage and disposal media for radioactive wastes is predicated upon the following properties of salt:

(1) Impermeability of Salt. Salt is impervious to the passage of water. Since salt goes so easily into solution, approximately 36 grams per 100 milliliters of water, we can reason that if water had been able to penetrate any of the salt formations, it would have left cavernous holes and caves similar to those developed in limestones by the passage of water through cracks and faults. Actually, we could expect even larger holes and caverns in salt than in limestone on the basis of the relative solubility of salt and dolomitic limestone: (35.7 gm compared to 0.032 gm per 100 ml of cold water) a ratio of 1100 to 1.

However, the salt has moved plastically to repair these cracks and faults and, therefore, only the surfaces of the deposits have come into contact with water. The plastic movement of salt is well illustrated above the potash mines of Carlsbad, New Mexico, where surface slumps of 5 to 7 feet have occurred without cracks appearing in the salt formation overlying the potash.

In salt mines some empirical tests have also shown the plastic movement of salt. At the Neu-Stassfurt Mine in Germany, at the turn of the century, measurements were made by placing fitted lead bars into holes 40 millimeters in diameter and 50 centimeters deep.[13] The bars at a depth of 1640 feet were jammed after a few months, those at 964 feet jammed after 2 years, and above 820 feet the holes remained open. Few of the salt mines in this country are below 1000 feet. Hence, there are few measurements of plastic flow under conditions likely to occur in mines. Most tests have been short-term tests, whereas in a disposal site we would be most concerned about the long-term

effects of moderate loads continuously applied. Other manifestations of the plasticity of rock salt are shown by the solid state physicists who cleave off the surface layers of salt crystals and then bend them into random shapes. Barnes also comments upon the plasticity of salt after the surface of the crystal has been wetted.(14) This accomplishes the same thing as the cleavage; the stressed planes are either annealed or removed.

A massive illustration of the plasticity of salt is shown in the salt domes of the Gulf Coast area. Here the salt has flowed plastically through weaknesses in the overburden to form large plugs of salt in and among the overlying sediments. This will be discussed in greater detail in the section on the geology of the salt formations.

There is some disagreement as to the impermeability of salt. Tests during the development of dissolved cavities in salt have shown that salt may be somewhat permeable. In these solution cavities, however, it is possible to have lenses of impurities and it may be through these layers, or along the interfaces, that the fluid moves. However, liquified petroleum gases (LPG) and gasoline have been stored in salt without losses and salt mines have been operating for over 100 years without evidence of seepage of water. Therefore, the wastes stored in salt should not reach nor contaminate surface or ground waters due to the fact that they will not come into contact with these waters.

(2) Wide Distribution of Salt. Salt is widely distributed, as shown in Figure 4.(15) Except for the Pacific Coast States, rock salt deposits are relatively close to all major population centers of the United States. The cost of shipping fuel elements to the fuel element reprocessing plant at the disposal site, or from the reprocessing plant to the disposal site, is a major cost in the over-all fuel recycling-waste disposal cycle, as high as 60 to 70

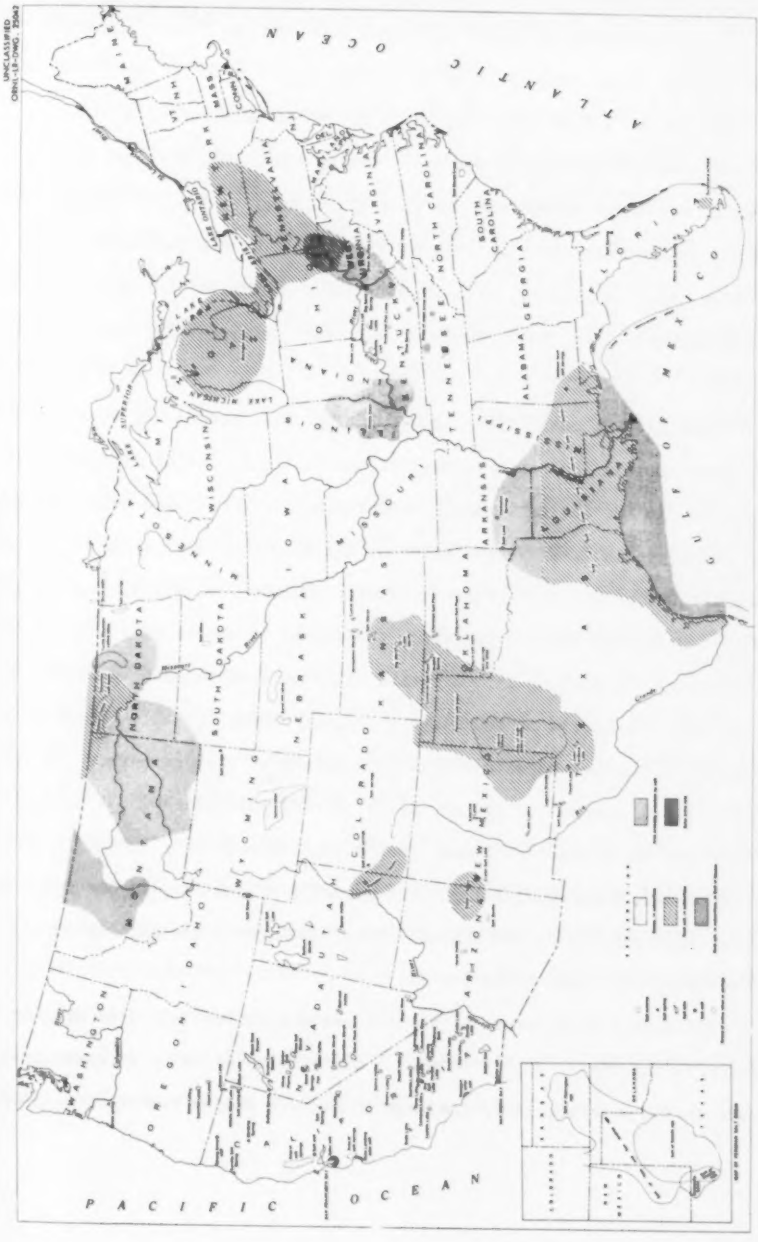

Fig. 4. Index Map of Salt Occurrences in the United States.

per cent.[16] Therefore, the short distances from nuclear power plants to the salt-disposal sites could be a major saving.

Halite, or sodium chloride, as it occurs in nature, varies to a marked extent depending upon its history. Having been usually formed by the concentration of saline waters and the precipitation of salts, the bedded deposits are interstratified with the other sediments of the saline waters - commonly clay, marl, sandstone, limestone, gypsum, anhydrite, dolomite, and shale. Consequently, the constituents of salt and its properties can vary quite widely.

There are two basic types of salt structures, bedded and dome. The salt formations in the northern part of the United States as typified by the deposits in the Salina formation underlying 100,000 square miles beneath New York, Pennsylvania, West Virginia, Ohio, and Michigan; the Permian formation underlying 120,000 square miles beneath Kansas, Colorado, Oklahoma, Texas, and New Mexico; and the Charles and Jefferson formations in the western part of the United States, underlying 15,000 square miles beneath Montana and North Dakota, are bedded deposits of sedimentary origin. There the salt occurs in beds of variable thicknesses with overlapping lenses of varying purity. A known stratigraphic succession of the layers of rock salt beds cannot be predicted to extend for distances of more than a few hundred feet, though it may extend many miles. Commercially important deposits of bedded salt are found in New York, Michigan, Ohio, and Kansas. They extend from the outcrop downward to depths of more than 5,000 feet.

Bedded salt also occurs in several western states. Salt of Permian age is found in the Sevier Valley in Utah. Salt, believed to be of Pennsylvanian age, occurs in the Colorado River drainage in eastern Utah and western Colorado,

the Paradox salt basin. The extent of the salt has not been fully determined but apparently is not less than 10,000 square miles.

In the southwest, salt occurs in the Delaware Basin of New Mexico and Texas. The margins of the area are known only approximately, but the total area underlain by salt may be as large as 70,000 square miles.

In northern Louisiana, southern Arkansas, and east Texas, bedded rock salt of Jurassic (or Permian) age has been reached in widely separated wells. The Eagle Mills (Louann) salt is seldom fully penetrated in wells, but thicknesses of 500 to 1500 feet are normal. It is estimated that this horizon underlies an area of 180,000 square miles on the Gulf Coast. Salt domes also rise from this formation.

The greatest number of salt domes in the United States occurs in the Gulf Coast section, though several salt anticlines and domes have also been found in southeastern Utah and southwestern Colorado. Here salt from older sedimentary formations has flowed upward through fractures and weaknesses in the overlying rocks (sp. gr. 2.6 vs. sp. gr. 2.2 for salt) to form plugs and domes in the overlying sediments. The diameter of the plug may be as much as 4 miles. The distance to the top of the domes varies from just below ground surface to depths greater than 10,000 feet.

The known salt domes are over 200 in number. The best known group of salt domes is the Five Islands of southern Louisiana, characterized by surface uplifts overlying the salt masses, and by the shallowness of the salt, less than 300 feet from the surface. At a depth of 1000 feet, these domes are a mile or more in diameter.

A more detailed description of the location of salt deposits is presented in the Appendix to reference 8.

At present the main interest in salt domes has been that of oil companies searching for oil along the salt barriers. The oil companies are also finding it highly profitable to store 1 billion gallons of LPG in cavities created by solution mining in salt domes. This is more than four times the volume of high-level radioactive wastes expected to be produced from now till the year 2000.

(3) Volume of Salt. The volume of salt in these domes in Texas and Louisiana alone is more than 3×10^{11} cubic feet at depths less than 1000 feet and more than 5×10^{12} cubic feet at depths less than 5000 feet,[17] or enough to provide the world's need of salt (at the present rate of consumption) for over 6000 years. These deposits, of course, are infinitesimal in comparison to the enormous amounts in the bedded deposits underlying wide geographic areas.

As of 1943, the estimated reserves of salt in the various states totaled 60,000 billion tons as shown in Figure 5.[18] In Figure 4 it is observed that large deposits of salt in North Dakota, Montana, Arizona, New Mexico, Arkansas, Mississippi, and Alabama, have been located since that time, and this estimate must, therefore, be increased. It is obvious that there is no shortage of salt. There is little likelihood that we will contaminate or render useless a valuable natural resource. There remains, of course, the salt in the ocean if that improbable event should occur. If all the salt in the ocean were recovered it would total 4 1/2 million cubic miles.

(4) Structural Properties of Salt. The strength of salt as a structural material is such that cavities 100 feet high, 100 feet wide, and 3000 feet long, can be opened at depths 700 feet below the ground surface and remain open without support of any kind. Figure 6, taken at the Kleer mine

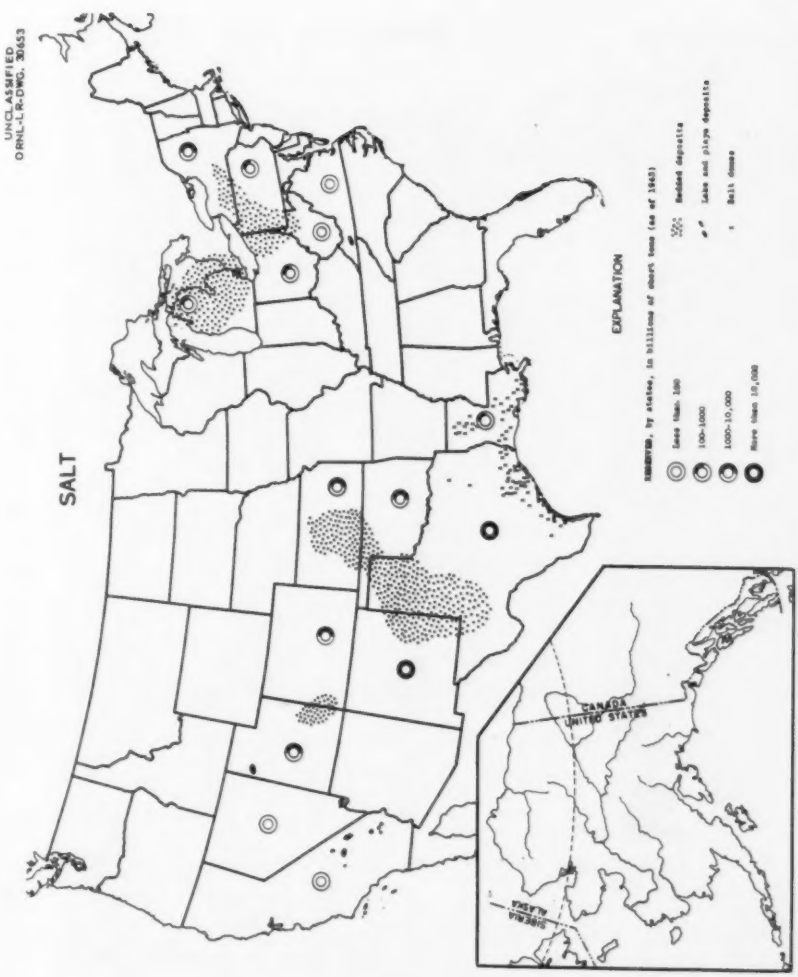

Fig. 5. Distribution of Estimated Salt Reserves of the United States as of 1943, by States.

Fig. 6.

of the Morton Salt Company at Grand Saline, Texas, shows the inside of a typical dome deposit mine. In Colombia, South America, there is even a cathedral inside a salt mine. The compressive strength of rock salt is about 5000 pounds per square inch. This compares with a compressive strength of 50,000 pounds per square inch for steel, 12,000 pounds per square inch for granite, and 5000 pounds per square inch for sandstone. As discussed earlier, the physical constants given for salt are a function of the history and composition of the specimen and the variations in samples can be quite striking.

(5) Thermal Conductivity of Salt. Salt has a high coefficient of thermal conductivity in comparison to other rocks as shown in Table III.

The higher coefficient of thermal conductivity, though low by comparison with the metals, will enable the heat generated by the fission product decay to be more quickly dissipated to the surrounding areas than would be possible in other rock media.

(6) Containment in Salt. One other advantage in the use of salt as a disposal medium for radioactive wastes is that radioactive materials still remain accessible if, at some future time, recovery should seem desirable. The value of some of the rarer elements produced by fission-product decay could help to pay for the disposal operations. Other disposal methods preclude the recovery of valuable by-products, except at extraordinary effort and expense. Glueckauf has estimated, at 1955 prices, that the resale of the decayed fission products - ruthenium, rhodium, palladium, and xenon - alone would amount to $180,000 per ton of U235 burned.(19)

(7) Economics of Developing Space in Salt. The cost of developing space in salt formations is low. Under average conditions solution cavities can be formed for 1 to 5 cents per gallon for capacities of 1 to 2 million

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TABLE III.
THERMAL CONDUCTIVITIES

MATERIAL	TEMPERATURE (°C)	Btu/(hr (sq ft) (°F/ft)
COPPER	100	222.8
IRON (99Fe 1 C)	100	26.1
BASALT	20	1.3
CONCRETE	20	0.5
EARTH'S CRUST, AVG.	20	1.0
FELDSPAR	20	1.4
GYPSUM	0	0.8
QUARTZ ∥ TO AXIS	100	5.2
⊥ TO AXIS	100	3.2
ROCK SALT	100	2.8
SANDSTONE	40	1.1
SOIL, DRY	20	0.1

gallons.[20] Larger cavities are cheaper. Wells are drilled to the rock salt and water pumped down to dissolve the salt, and then the brine is pumped back to the surface. Solution cavities, in addition to their lower cost, could handle liquid wastes and even some solid wastes. Excavated cavities are higher in price; normal costs range from 2[21] to 10 cents per gallon.[20] The structural qualities of salt allow modern excavating equipment to be used without regard to headroom considerations as is evident in Figure 6. Therefore, space can be easily and cheaply mined; in fact, so much so that the salt sells for as little as $3.00 per ton at the mine.[22] Mined cavities allow access to the disposal area, give greater flexibility in the use of the space, allow the cavity to be placed in the soundest formations, allow inspection prior to use, and eventually would make recovery of the decayed fission products easier. This would be true for both solid and liquid wastes. Table IV shows the relative advantages of various underground reservoirs for petroleum products.[23] The same relative ratings should hold for underground storage of radioactive wastes.

(8) Seismic Disturbances in Salt Areas. Salt formations are located in areas of low seismicity; therefore, we may expect only small and infrequent disturbances to the structure.

Figure 7 shows that in the areas in which salt formations are located, few, if any, earthquakes of appreciable force have occurred.[24]

Problems in the Disposal of Waste in Salt Cavities

Despite all the favorable factors, the storage of radioactive wastes in salt has not yet taken place because the conditions involved in waste storage will be different than those ordinarily encountered in LPG storage or mining of salt. The degree to which the desired structural properties of salt are

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TABLE IV.

COMPARATIVE RATINGS OF UNDERGROUND RESERVOIRS FOR PETROLEUM STORAGE

Type of Reservoir	Cost of Construction	Lack of Contaminants	Cost of Injection and Recovery	Total of Factor Ratings	Economic Preference Rating
Solution cavity in salt dome	1	1	2	4	I
Solution cavity in bedded salt	1	2	2	5	II
Mined cavity in shale or salt	3	2	1	6	III
Mined cavity in hard rock	5	1	1	7	IV
Natural trap reservoir in porous rocks	1	3	4	8	V
Abandoned mine or natural cavern (would be rated high if easy to seal)	3	5	1	9	VI

Underground storage considered impractical in areas of volcanic rocks, mineralization, extremely compl structure, or excessive general permeability.

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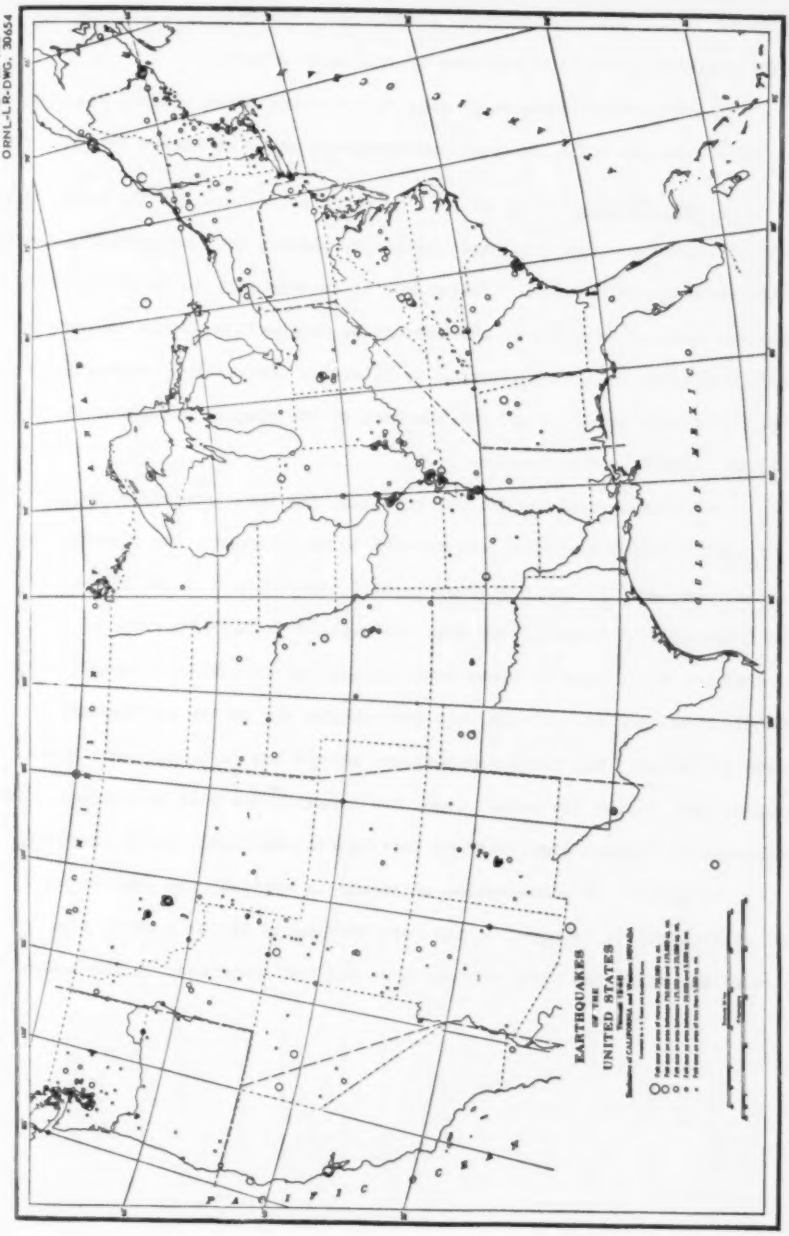
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EARTHQUAKES
OF THE
UNITED STATES
GULF OF MEXICO
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Fig. 7.

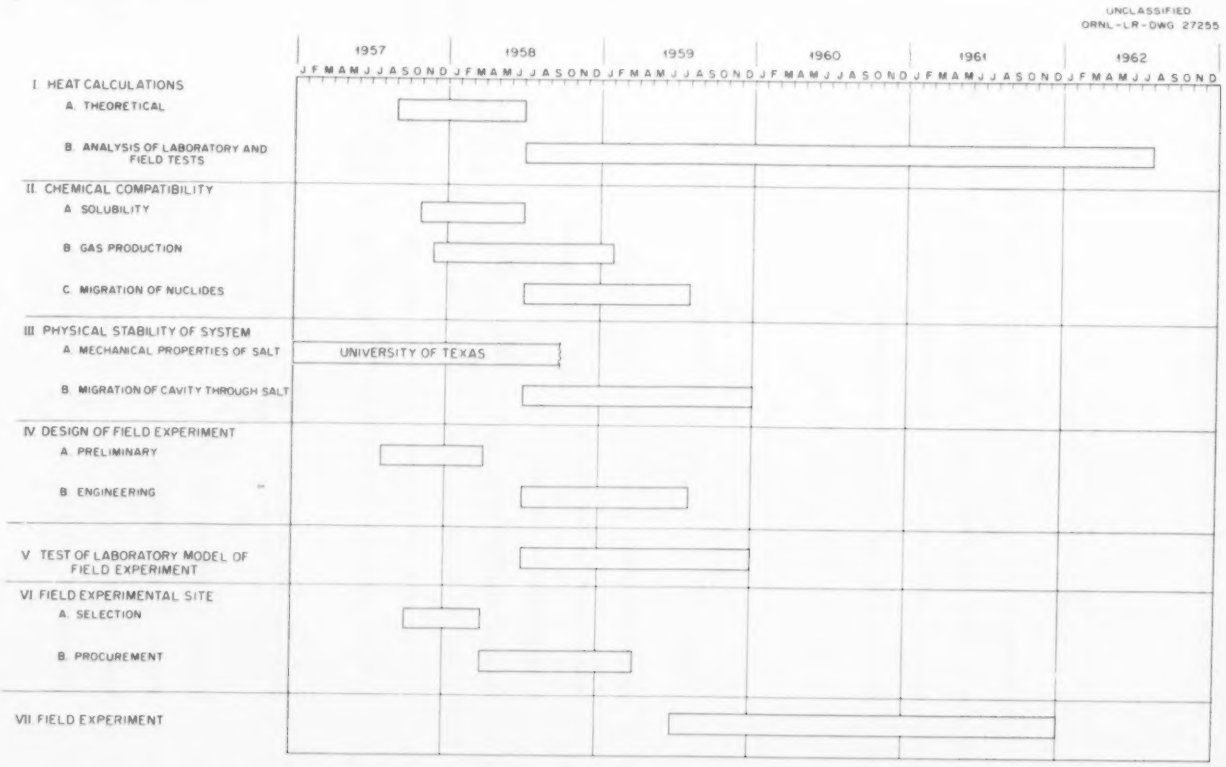
affected by liquid wastes, pressure, temperature, and radiation will determine the role of salt in the disposal of radioactive wastes.

A seven-point research program to determine these effects has been initiated at the Oak Ridge National Laboratory as shown in Figure 8.

I Heat Calculations. As will be shown in Section III the plasticity of salt and therefore the structural integrity changes to a considerable degree with temperature and pressure. Therefore, it is necessary to determine the temperature rise in salt due to the storage of radioactive wastes in cavities in salt formations. Initial attempts to determine the maximum temperature possible in a salt cavity have been confined to the simpler geometric cases - flat plane, sphere, and a cylinder.

To further simplify the calculations, the heat generating source (tne decaying fission products) was assumed to be constant. The thermal conductivity, the density, and the specific heat were assumed to be independent of the temperature. Finally, the heat was assumed to be dissipated by conduction alone, no changes in state were allowed, no convective currents were permitted in the wastes, and the salt surrounding the cavity was assumed infinite in extent. All these assumptions, except the last one, are on the safe side, that is, in the actual case, the temperatures will be reduced. The solution can be further simplified by solving for the steady state case only.

If the law of conservation of energy is applied, the heat in the sphere after time, t, is equal to the heat originally in the sphere, plus the heat entering, minus the heat leaving, plus the heat generated in the sphere or: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I. HEAT CALCULATIONS
A. THEORETICAL
B. ANALYSIS OF LABORATORY AND FIELD TESTS
II. CHEMICAL COMPATIBILITY
A. SOLUBILITY
B. GAS PRODUCTION
C. MIGRATION OF NUCLIDES
III. PHYSICAL STABILITY OF SYSTEM
A. MECHANICAL PROPERTIES OF SALT
UNIVERSITY OF TEXAS
B. MIGRATION OF CAVITY THROUGH SALT
IV. DESIGN OF FIELD EXPERIMENT
A. PRELIMINARY
B. ENGINEERING
V. TEST OF LABORATORY MODEL OF FIELD EXPERIMENT
VI. FIELD EXPERIMENTAL SITE
A. SELECTION
B. PROCUREMENT
VII. FIELD EXPERIMENT

Fig. 8.

And if the law is also applied outside the sphere the equation is:

$$\frac{1}{r^2} \frac{d}{dr} \left(r^2 \frac{dT_s}{dr}\right) = 0$$

Furthermore, if the law of continuity is applied at the boundary of the sphere so that the temperature at the boundary is the same inside and outside the sphere, and if the heat flux at the boundary of sphere is the same inside and outside the sphere, mathematically:

$$T_w = T_s$$

$$\text{and } K_s \frac{T_s}{r} = K_w \frac{T_w}{r}$$

where r = radial distance from center of sphere, Ft

T_w = temperature in the waste solution, ^oF

T_s = temperature in the salt, ^oF

K_s = thermal conductivity, salt $\frac{BTU}{Hr} \frac{Ft}{Ft^2\ ^oF} = \frac{BTU}{HrFt\ ^oF}$

K_w = thermal conductivity, waste $\frac{BTU}{Hr} \frac{Ft}{Ft^2\ ^oF} = \frac{BTU}{HrFt\ ^oF}$

A_o = heat generating source, $\frac{BTU}{Hr\ Ft^3}$

a = radius of sphere, Ft

b = radius of salt media, Ft

t = time, hours

k = thermal diffusivity = $\frac{K}{c\rho} = Ft^2/Hr$

c = specific heat, $\frac{BTU}{lb\ ^{\circ}F}$

ρ = density, lbs/Ft^3

$C = c\rho$ = specific heat per unit volume $\frac{BTU}{^{\circ}F\ Ft^3}$

$$\operatorname{erfc} x = 1 - \operatorname{erf} x = \frac{2}{\sqrt{\pi}} \int_x^{\infty} e^{-\beta^2}\, d\beta$$

$$i^n \operatorname{erfc} x = \int_x^{\infty} i^{n-1} \operatorname{erfc} \beta\, d\beta \qquad n = 1, 2, \ldots,$$

β^2 = variable of integration = $\frac{a^2}{4k(\tau - t)}$ and $\frac{a^2}{k(\tau - t)}$

λ = decay constant, $\frac{1}{hours}$

τ = a fixed value of t, hours

If it is further stipulated that the temperature is zero at the outside boundary of the salt and that the rate of change of temperature with respect to the radius at zero radius is zero, then the solution of the steady state temperature in the waste is equal to:

$$T_w = \frac{A_o}{3} \left[\frac{a^2}{K_s} - \frac{a^3}{K_s b} + \frac{a^2}{2K_w} - \frac{r^2}{2K_w} \right]$$

and if b is assumed infinite and r = 0

$$T_w = \frac{A_o}{3} \left[\frac{a^2}{K_s} + \frac{a^2}{2K_w} \right]$$

and if $K_w = K_s$, $T_w = A_o \frac{a^2}{2K_s}$ and if $K_w \longrightarrow \infty$ (that is the waste is considered infinitely well stirred) then:

$$T_w = \frac{A_o a^2}{3K_s}$$

If we limit the maximum temperatures to be permitted at the edge of the sphere, then the maximum concentrations of heat generating sources (fission products) allowed in spheres of various sizes are shown in Figure 9.[25] If it is assumed that there is thermal convection in the wastes, all other conditions remaining the same, then the permitted concentrations of fission products in spheres are shown in Figure 10.[25]

Professor Birch of Harvard University has solved analytically the transient problem for a flat plate and a sphere with a decaying heat generating source for the case of equal conductivity and specific heat for the waste and the salt and for the case of infinite conductivity of the waste or a well-stirred waste.[26] The former is the same as the correct solution for the equations on page 287 and page 288 of Carslaw and Jaeger or:[27]

$$0 \leqslant r < a \qquad T(r,t) = \frac{A_o t}{C} \left[1 + \frac{2a}{r} \left(i^2 \operatorname{erfc} \frac{a + r}{\sqrt{4Kt}} - i^2 \operatorname{erfc} \frac{a - r}{\sqrt{4Kt}} \right) \right.$$

$$\left. + 2 \frac{\sqrt{4Kt}}{r} \left(i^3 \operatorname{erfc} \frac{a + r}{\sqrt{4Kt}} - i^3 \operatorname{erfc} \frac{a - r}{\sqrt{4Kt}} \right) \right]$$

and for $r > a$

HEAT GENERATING SOURCE PER UNIT VOLUME (Btu / hr / gal)

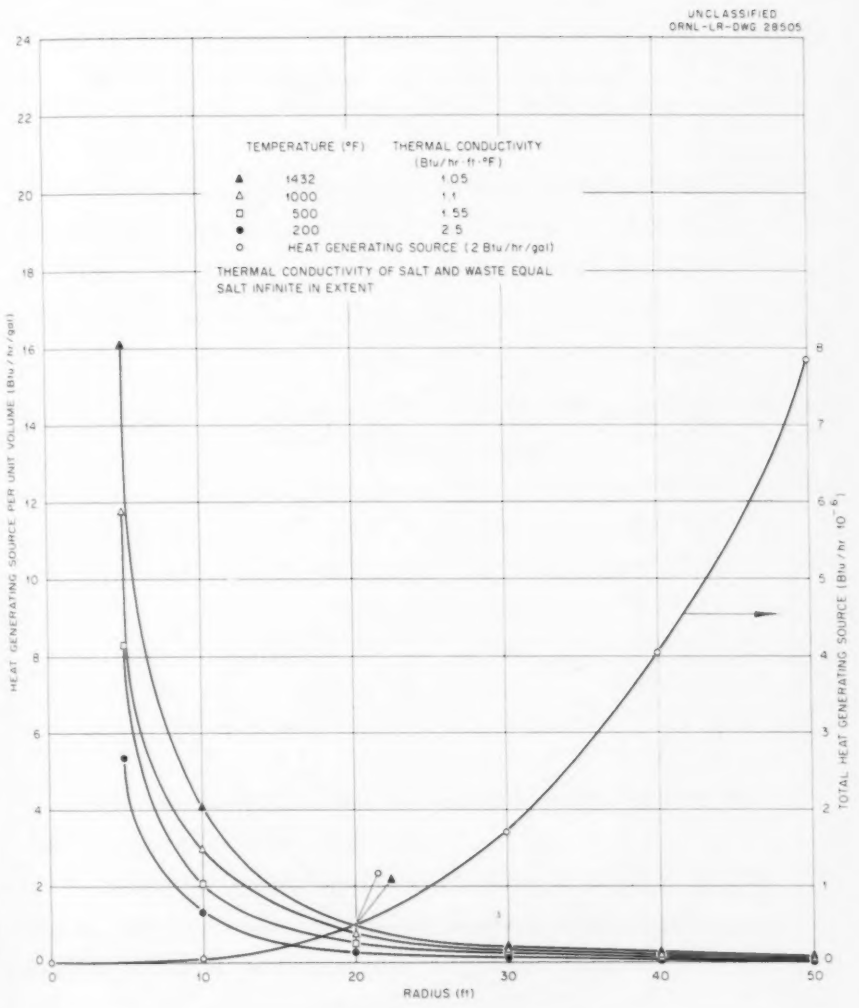

Fig. 9. Heat Source Required to Achieve Steady State Temperature in a Sphere.

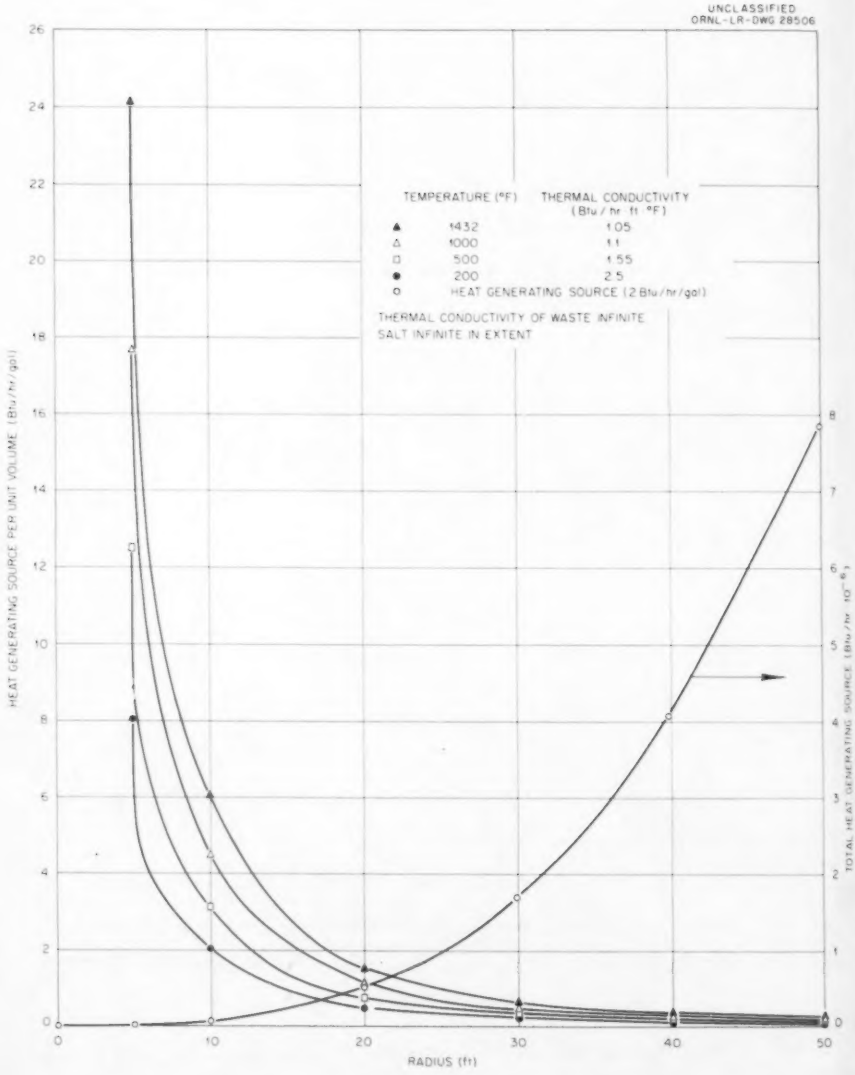

Fig. 10. Heat Source Required to Achieve Steady State Temperature in a Sphere.

$$T(r,t) = \frac{2Aat}{Cr}\left[i^2 \operatorname{erfc} \frac{r-a}{\sqrt{4\kappa t}} + i^2 \operatorname{erfc} \frac{r+a}{\sqrt{4\kappa t}} + \frac{4\kappa t}{a}\left(i^3 \operatorname{erfc} \frac{r+a}{\sqrt{4\kappa t}} - i^3 \operatorname{erfc} \frac{r-a}{\sqrt{4\kappa t}}\right)\right]$$

These, of course, reduce to the results obtained by Julian Crowell when the steady state is reached (i.e., when $t \longrightarrow \infty$). When the heat generating source is assumed to decay exponentially, Birch derives the following solution:

$$r = 0;\quad T(0,t) = \frac{A_o a^2}{2Ck}\left\{ e^{-\lambda t} \int_{a/\sqrt{4\kappa t}}^{\infty} e^{\lambda a^2/4\kappa\beta^2}\left(\operatorname{erf}\beta - \frac{2\beta}{\sqrt{\pi}} e^{-\beta^2}\right)\frac{d\beta}{\beta^3}\right\}$$

and for $r = a$

$$T(a,t) = \frac{A_o a^2}{3Ck}\left\{ 3e^{-\lambda t} \int_{a/\sqrt{\kappa t}}^{\infty} e^{\lambda a^2/\kappa\beta^2}\left(\operatorname{erf}\beta - \frac{2}{\beta\sqrt{\pi}} 1 - e^{-\beta^2}\right)\frac{d\beta}{\beta^3}\right\}$$

The functions in the last two equations are not tabulated and, therefore, must be integrated numerically.

Numerical integration by hand of a number of cases can be quite tedious. However, Professor Schecter of University of Texas working independently has solved the above equations, assuming a decay rate varying with

time and had it programmed on the ORACLE (digital computer at ORNL). His results give the maximum temperature for any waste concentration in any size sphere for 2, 6, and 10-year cooled wastes.[28] They do not give, however, the time-space distribution of the temperature. The proper size of a test cavern which would give temperature results in a reasonable time was determined by the temperature, time, and space relationship of a 6-year cooled waste, (2 BTU/hr/gal) with a constant heat generating source (for ease in computation).

Based upon Professor Birch's statement, "that heating the liquid some hundred degrees (Centigrade) or so would not produce drastic physical changes, and that plastic deformation of shale or evaporite layers, if sufficiently thick might continue to provide a tight container,"[26] it was decided to limit the temperature rise at the edge of the sphere to 200° F. This would also eliminate any necessary corrections due to changes in state as the temperature is kept below the boiling point of the wastes. Using Crowell's curves, it was found that for 2 BTU/hr/gal waste that a sphere, 10 feet in radius, would not reach a temperature greater than 200° F for the case where the thermal conductivity of the waste is infinite.

Based on Birch's and Goldenberg's equations,[29] the time-space distribution of the temperature for a 20-foot diameter sphere with a constant heat generating source and thermal conductivity of the waste equal to the thermal conductivity of the salt was found to be that shown in Figure 11.

Work is continuing now to determine the time-space distribution of temperature for the case where the heat generating source is decaying as a function of time and where the thermal conductivity in the sphere is equal to infinity.

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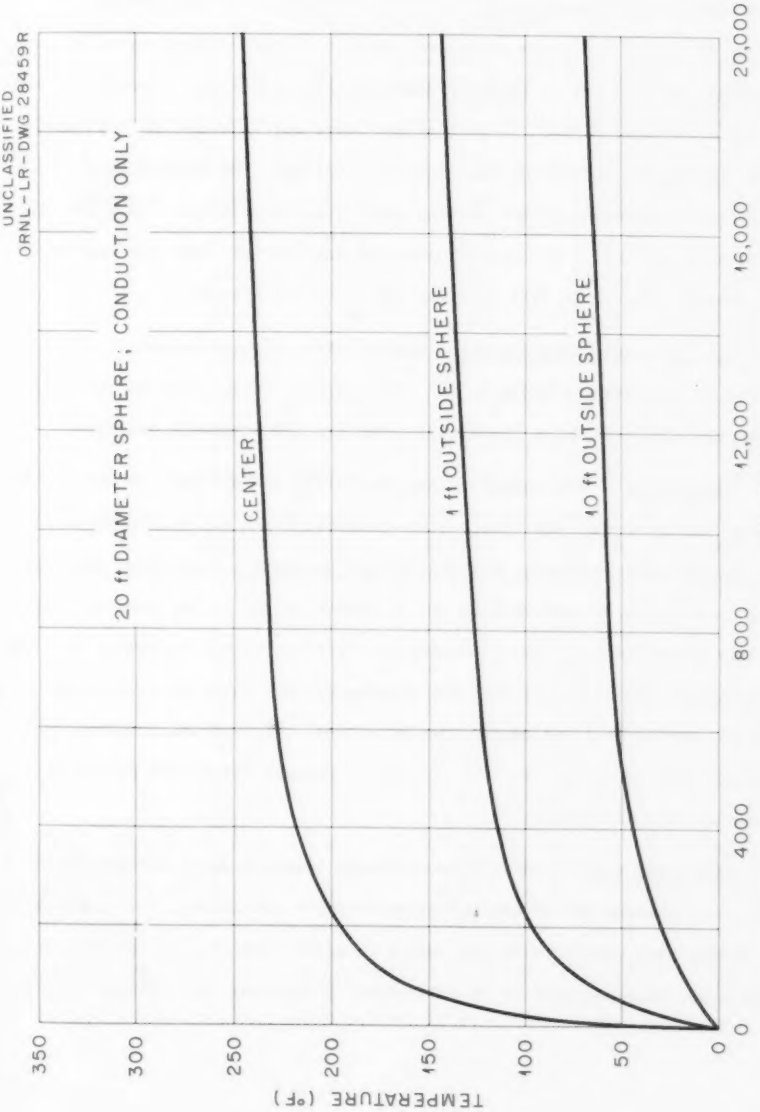

Fig. 11. Time-Space Temperature Distribution of Wastes Stored in Salt Cavities.

In all the cases treated above, the heat has been dissipated solely by conduction. Preliminary studies by G. W. T. Kearsley of the Chemical Technology Division on the use of natural convection for cooling wastes in salt indicate that it may be feasible under certain conditions. Further studies of the use of natural convection systems are under way. Though not restricting ourselves to natural convection and conduction systems, the desirability of such a system is apparent if the cooling periods are considered. If a mechanical cooling system should be required over the long periods that the wastes must be stored, storage in salt cavities might be uneconomical.

II. Chemical Compatibility. Typical chemical composition of rock salt deposits are given in Table V.[30] The chemical composition may be more complex than the simple formulas cited, but these are the compounds analyzed.

Solubility. The studies of the solubility of NaCl and its associated impurities, $CaCl_2$, $MgCl_2$, and $CaSO_4$, with synthetic Purex and acid aluminum nitrate non-radioactive wastes, neutralized and untreated, have shown that the salts are soluble in the neutralized and untreated wastes to the same degree, about 50 to 90 per cent of the solubility in distilled water, depending upon the concentration of the wastes, except for calcium sulfate which is five to ten times as soluble in acid and neutralized solutions. The results obtained by E. Eastwood are shown in Table VI. The solubility changes due to the use of radioactive wastes will be studied.

Gas Production. Chlorine and nitrogen compound gases are generated when the unneutralized wastes and the chloride salts are mixed. The highly acid nitrate wastes upon contact with the sodium chloride oxidizes the chloride and generates a gas which appears to be predominantly chlorine and nitrogen compounds.

TABLE V

COMPOSITION OF ROCK SALT

	Retsof New York	Detroit Michigan	Avery Island Louisiana	Hutchinson Kansas	Natrium West Virginia
Moisture %	0.015	0.114	0.019	0.670	0.350
Water insoluble %	0.962	0.756	0.711	1.407	1.070
$CaSO_4$	0.743	0.872	0.225	0.857	1.600
$CaCl_2$	0.006	0.053	0.010	0.034	0.230
$MgCl_2$	0.002	0.024	0.009	0.066	-----
$MgSO_4$	-----	-----	-----	-----	-----
Na_2SO_4	-----	-----	-----	-----	-----
K Cl	-----	-----	-----	-----	0.310
NaCl	98.262	98.181	99.026	96.970	96.440
	99.990	100.000	100.000	100.04	100.00

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SOLUBILITY (g/100 ml)

Waste Type	Rock Salt Grand Saline			Sodium Chloride Analytical Reagent Grade			Calcium Sulfate Analytical Reagent Grade			Magnesium Chloride Analytical Reagent Grade			Calcium Chloride Analytical Reagent Grade		
	2°C	23°C	70°C	2°C	23°C	75°C	2°C	23°C	70°C	2°C	22°C	70°C	2°C	23°C	74°C
Distilled water	32.2	32.5	31.6	31.5	32.5	31.7	0.21	0.21	0.20	44.0	45.5	48.3	51.8*	51.1*	75.2*
2 M HNO_3	26.3	26.9	26.5	25.3	26.9	25.8	1.59	1.65	1.60	41.4	42.7	45.9	49.2*	40.5*	57.2*
4.5 M HNO_3	19.3	20.5	19.1	21.3	20.8	22.4	1.76	1.71	1.83	36.6	40.5	46.3	60.0*	56.1*	63.4*
7.0 M HNO_3	18.1	18.1	17.6	18.2	19.0	17.4	1.93	2.06	2.64	32.7	38.3	41.1	55.4*	42.8*	55.9*
0.10 M HNO_3 1.6 M $Al(NO_3)_3$	23.4	23.1	23.1	22.8	23.4	23.6	1.94	2.03	2.06	36.4	40.3	41.5	42.0*	43.2*	48.3*
0.16 M HNO_3 1.6 M $Al(NO_3)_3$	21.5	22.2	22.0	21.6	22.5	22.1	2.09	2.22	2.11	37.8	41.2	39.6	41.6*	42.7*	41.7*
0.22 M HNO_3 1.6 M $Al(NO_3)_3$	23.2	23.1	22.9	21.3	22.8	21.8	2.03	2.00	2.21	35.2	39.3	40.5	41.9*	40.5*	44.0*

*Formed Gel – Difficult to sample.

SOLUBILITY (g/100 ml)

pH7 With NaOH	Rock Salt Grand Saline			Sodium Chloride Analytical Reagent Grade			Calcium Sulfate Analytical Reagent Grade			Magnesium Chloride Analytical Reagent Grade			Calcium Chloride Analytical Reagent Grade		
	2°C	23°C	88°C	2°C	23°C	85°C	2°C	23°C	85°C	2°C	23°C	80°C	2°C	23°C	85°C
Distilled water	31.4	32.9	32.7	31.9	32.3	32.4	0.19	0.20	0.19	43.8	45.4	47.6	49.4*	58.8*	71.2*
2 M HNO_3	27.6	28.3	27.6	27.9	28.3	28.8	0.69	0.72	0.69	41.2	42.4	46.0	45.6*	53.6*	62.8*
4.5 M HNO_3	24.9	25.0	24.1	24.4	24.5	24.7	0.90	0.90	0.80	38.8	42.7	44.0	*	52.4*	63.0*
7.0 M HNO_3	21.5	21.2	21.6	21.9	22.3	24.0	0.90	0.89	0.83	38.3	39.1	40.0	*	54.2*	65.4*
0.10 M HNO_3 1.6 M $Al(NO_3)_3$	18.0*	20.2*	18.0*	20.4*	24.3*	19.7*	3.49*	3.36*	2.92*	38.4*	42.2*	42.7*	*	74.7*	69.8*
0.16 M HNO_3 1.6 M $Al(NO_3)_3$	19.0*	20.4*	23.1*	15.9*	22.1*	17.9*	3.40*	3.42*	3.25*	38.6*	42.4*	42.8*	*	68.0*	69.2*
0.22 M HNO_3 1.6 M $Al(NO_3)_3$	18.3*	20.7*	18.5*	21.3*	23.5*	20.3*	3.38*	3.25*	2.95*	38.2*	41.6*	42.8*	*	72.6*	66.2*

*Formed Gel – Difficult to sample.

TABLE VI. SOLUBILITY OF SALTS IN WASTE SOLUTIONS.

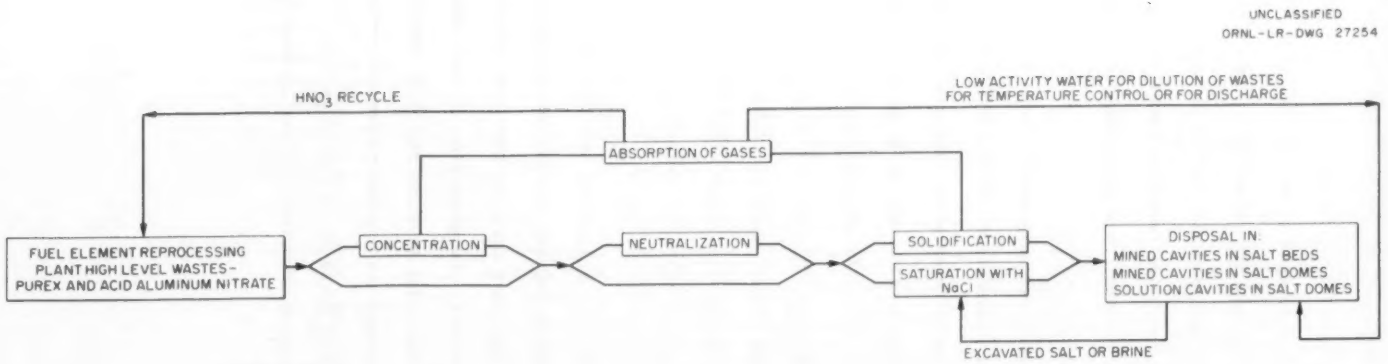
The rate, volume, and composition of the gas under the varied conditions outlined above are presently under study at Oak Ridge. The effect of radiation upon such a liquid system is to tend to hasten the rate of the reaction and to carry the reaction further as the radiation induces V centers in the sodium chloride which traps holes and upon dissolution oxidizes the chlorides to chlorine.[31] (V centers are holes trapped at positive ion vacancies or aggregates of vacancies. V centers are also formed when the crystals are exposed to the vapor of the halogen.)

If the wastes are previously neutralized, the production of gas is negligible. The effect of radioactive neutralized wastes on salt will be studied.

The results of these experiments indicate that the wastes will have to be neutralized and saturated with salt prior to disposal in salt cavities. The generalized flow sheet will then look something like Figure 12.

Migration of Nuclides. Though salt is generally considered to be impermeable and the dryness of salt mines is convincing testimony to that effect, it is known that water molecules will migrate in rock salt to some extent.[14] The question, therefore, arises as to the permeability of rock salt for specific radionuclides.

The migration of radionuclides in salt has been particularly difficult to determine in the laboratory because undisturbed samples cannot be obtained and tests of the samples used have shown in most cases that the samples were fractured in the mining process. Large samples of rock salt have been obtained and the migration of radionuclides in these blocks at various temperatures will be studied. It is obvious that the typical properties of the salt in the laboratory are not the same as they would be in a mine because of the release of the overburden pressure. Therefore, the tests in the laboratory can only prove that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HNO_3 RECYCLE
LOW ACTIVITY WATER FOR DILUTION OF WASTES FOR TEMPERATURE CONTROL OR FOR DISCHARGE
ABSORPTION OF GASES
FUEL ELEMENT REPROCESSING PLANT HIGH LEVEL WASTES - PUREX AND ACID ALUMINUM NITRATE
CONCENTRATION
NEUTRALIZATION
SOLIDIFICATION
SATURATION WITH NaCl
DISPOSAL IN:
MINED CAVITIES IN SALT BEDS
MINED CAVITIES IN SALT DOMES
SOLUTION CAVITIES IN SALT DOMES
EXCAVATED SALT OR BRINE

Fig. 12. Disposal of Solid and Liquid Radioactive Wastes in Salt Formations.

salt is impermeable, but not disprove it as the relief of the overburden pressure and the possible fracturing of the sample can only lead to a more permeable sample.

III. Physical Stability of the System. The effect of radiation upon NaCl is well known for single crystals. The solid state physicists have found the alkali halides particularly applicable to their studies of crystals, and hence there is a large amount of information available. It appears that there is a finite limit to the radiation damage possible. An upper limit to the concentration of F* and V centers is set by the fact that when an F center gets close enough to the V center, the electron in the F center can tunnel through to the V center and annihilate the positive hole. The maximum degree of decomposition of the salt is indicated to be of the order of one-tenth of 1 per cent.(32)

The structural properties, however, can change to a greater degree. The effect of x-ray irradiation upon rock salt has been to increase its hardness, tensile strength, and modulus of elasticity.(33)

This has been attributed to the pinning of dislocations by vacancies which condense near dislocations in the crystals.(34) The effect of radiation upon salt aggregates is being studied at the University of Texas with samples irradiated at ORNL.

Mechanical Properties of Salt. Most of the experiments to determine mechanical properties of salt have been performed on single and synthetic crystals. Therefore, the results do not have a great deal of pertinence to the problems encountered in salt cavities. These values can vary considerably for rock

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*F centers - electrons trapped at halogen ion vacancies or when the crystal is exposed to the vapor of the alkali metal.

crystals and aggregates. The values quoted below are for rock salt aggregations of crystals rather than for synthetic and single crystals. Most of this work has been performed by M. Stamatiu, a Roumanian mining engineer, in the early 1930's.(35) The only comparable work on aggregations of crystals is that currently under way at the University of Texas under Professor Gloyna. Table VII gives the mechanical properties of three types of rock salt found in the mines at Slanic-Prahova, Roumania. Table VIII gives the chemical composition of the three types of salt for comparison with American rock outlined in Table V. Since the chemical compositions are very similar we may use these values as a guide for the design of the cavities. For the final designs we shall have to take a number of samples from the salt to be excavated. The work of Stamatiu has shown that even in the same bed, the properties can vary considerably.

The increase in plasticity in rock salt crystals with temperature and pressure is quite rapid as shown in Figure 13.(36) It is apparent then that the temperature and pressures in the waste-disposal cavity will have to be kept below those temperatures and pressures which would cause enough plastic flow to make the structure unsafe. The plastic flow of rock salt aggregate is presently being measured at the University of Texas. It appears that as long as the temperature is kept below 200° C and a pressure of 100 g/mm^2 there will be no short-term plastic deformation in the mine. (1 gram per millimeter squared is approximately one-tenth of an atmosphere.)

Migration of the Waste Storage Cavity Through the Salt. A major deterrent to the storage of liquid radioactive wastes in salt cavities could develop if the heated and evaporating wastes were to condense upon a salt roof, dissolve the salt, and fall back to the wastes. This process, if continued for a period

TABLE VII

PHYSICAL PROPERTIES OF ROCK SALT OF ROUMANIA

	Pounds per Square Inch			
	White salt	Mottled salt	Gray salt	Average for samples from 5 mines
Compression	4720	5260	5320	4680
Tension	132	231	398	142
Bending	409	365	772	425
Shear	328	251	355	355
Unit Compression at Rupture	3.9%		4.14%	0.5 - 4.0
Poisson's Ratio				0.2 - 0.5
Density lbs/Ft^3				122 - 125
Hardness, Mohs Scale				2 - 3
Modulus of Elasticity				$.085 - .284 \times 10^6$

In general, one may say that rock salt behaves as a good concrete.

TABLE VIII

MEAN CHEMICAL COMPOSITION OF ROCK SALT OF SLANIC

Chemical composition	White Salt	Mottled Salt	Gray Salt
$NaCl$	99.6612	99.0036	98.5498
Na_2SO_4	-----	-----	0.5658
$MgCl_2$	0.0966	0.0800	-----
$MgSO_4$	-----	-----	0.1031
$CaCl_2$	0.0114	0.0978	-----
$CaSO_4$	0.0177	0.6327	0.7212
H_2O	0.2924	0.1768	0.0835
Insolubles	0.0080	0.0460	0.0520
Totals	100.0873	100.0369	100.0754

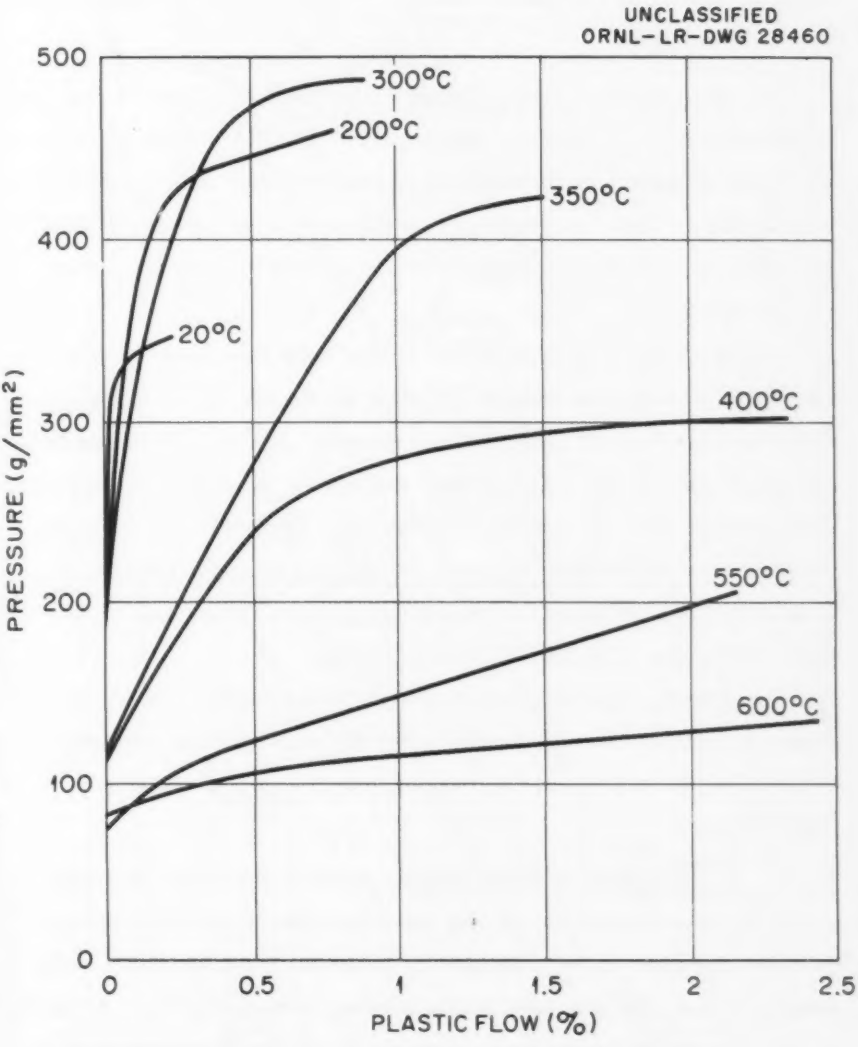

Fig. 13. Plastic Flow Curve for Natural Rock Salt.

of time, could progress through the salt formation and perhaps eventually cause the structural collapse of the mine. The possibility of this occurring will be investigated.

IV. Design of Field Experiment. The problems related to disposing of radioactive liquid wastes in salt formations have been briefly outlined in the preceding pages. In storing solid radioactive wastes the problems are considerably reduced. The only major problem then is the dissipation of the heat generated and the de-entrainment of the aerosols from the convective cooling stream.

The disposal of solid wastes in salt mines offers many advantages and may be the safest and cheapest method if the transformation of the liquid wastes to solid form can be economically achieved. One major advantage in the storage of solid wastes in salt is that they are not required to be nonleachable since no water will come into contact with them. Another advantage is that if the solid forms are properly designed, cooling will be easier and cheaper than cooling of liquid wastes. Cinder blocks might be one design, easy to cast, large surface area to volume, and easy to stack.

However, since there are at present no large amounts of reactor fuel element reprocessing wastes converted to solid form, the field experiment has been designed to test the safety, economy, and efficiency of the disposal of liquid wastes.

A Preliminary Conceptual Design. A preliminary conceptual design of what an operating facility in salt might look like is given in Figure 14. The field experiment is being designed to answer the questions raised in the paragraphs above with reference to this proposed conceptual design. Primarily the test will answer the technical questions of how good an approximation to

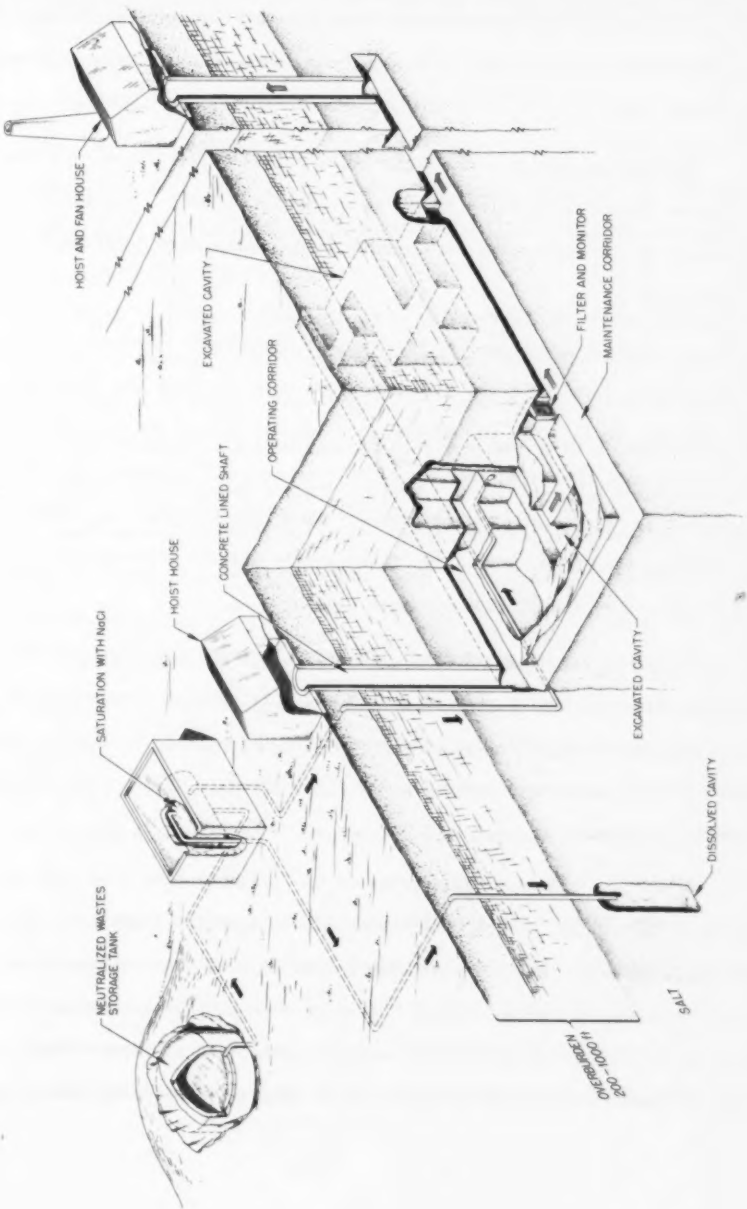

Fig. 14. Disposal of Radioactive Wastes in Salt Formations.

the physical model has our mathematical model been and how representative of field conditions have our tests in chemical compatibility and physical stability of the system been.

Engineering Design. The engineering design of the field experiment will commence after the location of the experiment is chosen. The design will be influenced by the concurrent experiments on the laboratory model of the field experiment.

V. Test of Laboratory Model of Field Experiment. Special rock salt samples of 4.5 and 27 cubic feet have been obtained from the Kleer Mine at Grand Saline, Texas. The blocks were drilled out rather than blasted and so should have less fractures than the usual samples, but possibly more than the rock salt in place. The temperature distribution, the chemical compatibility, and the structural stability of the cavity will be observed as the wastes are placed into cavities drilled in these blocks.

VI. Field Experimental Site. In order to solve the problems relating to the feasibility of the storage of liquid and solid radioactive wastes in dissolved and excavated cavities, pilot-scale operations will have to be carried out in the field. An active search is now in progress to obtain a field test site for our purposes.

Selection. The desirable criteria in picking a test site are: (1) suitable formation - geology well known with adequate thickness of salt, minimum depth to salt, free of extraneous shafts or core holes, homogeneous salt beds or domes; (2) accessibility - near paved roads, railroads, water transportation, living accommodations, skilled labor, and sufficient water; also near to Hanford, Savannah River, or Idaho Chemical Processing Plant, so

as to be able to obtain radioactive wastes economically, but not to have more weight than a location near the center of the future nuclear power complex; (3) removed from large population centers and active development of natural resources; (4) on Government-owned land if possible as this would make the property available and ease the problem of the supervision of the site for the period necessary; (5) suitable for a permanent disposal facility if the tests should prove to be successful.

There is general agreement that there should first be some tests using synthetic wastes and simulated heat sources followed by tests using hot wastes. In order to expedite the cold tests it would be desirable to run the tests in space presently available. It does not seem likely that hot tests would be permitted in mines still operating. Accordingly, a survey was made of the mines presently operating in the United States. The fourteen mines operating are located as follows: New York, 2; Michigan, 1; Kansas, 3; Louisiana, 4; Texas, 2; and Utah, 2.

On the basis of the desirable criteria, the mines in Louisiana and Texas can be ruled out of consideration, as they are too far from the center of the growth of the nuclear power complex where the first commercial fuel element reprocessing plant is likely to be located. Even for the test facility they are remote from Hanford, Idaho Falls, and Savannah River, the present sources of wastes.

An investigation into the possibilities of radioactive waste storage in salt formations in the northeastern United States has been prepared for us by the Geotechnical Corporation.[37] The report details the salt formations and mines in the area of the United States, bounded on the north by Canada, on the east by the Atlantic Ocean, and on the west by Montana, Wyoming, and Colorado,

and on the south by Oklahoma, Arkansas, Tennessee, and North Carolina.

The one operating mine in Michigan underlies the city of Detroit and would not be suitable for public relations reasons alone. Also the beds are relatively thin and deep lying there.

The mines in the states of New York and Kansas might possibly be used. It seems unlikely, however, that an operating mine will be available even for cold tests as the demands we would make upon the shaft and mining equipment would constitute a hindrance to the usual operations. The only stand-by mine reportedly in good condition is the Carey mine in the town of Lyons, Kansas. All the equipment, including the hoist, have been removed. Therefore, the only savings possible, if this mine could be obtained, would be the cost of the shaft minus the cost of the land and the cost of the mine. The existing space in the mine probably could not be used directly for liquid wastes, but might possibly be used for the storage of solid wastes. Therefore, new space, specifically designed for the storage of liquid waste, would have to be excavated.

The mines in Utah are quite small; they have mined out less than 100,000 cubic feet. Therefore, it does not seem possible to use these mines.

If none of the existing mines were available and suitable, it might be necessary to develop our own test space. Then according to the criteria outlined on the previous page, a site in Ohio along Lake Erie or in Kansas around Hutchinson, might seem most desirable. The depth to salt at Lake Erie is about 2000 feet versus 450 feet at Hutchinson. The thickness of the beds at Lake Erie is about 60 to 70 feet versus 220 feet at Hutchinson. The location on the Great Lakes is nearer the center of the likely nuclear power plants and has deep water transportation available to it. The hazards involved in water transportation, however, may negate this advantage. The Lake area is a more highly

developed and populated area and, consequently, it might be more difficult from a public relations view point to establish a waste disposal facility there. The consequences of a failure in a well-developed area such as one on the Great Lakes are, of course, much greater than those of a failure in the sparsely settled farming country of Kansas.

The greater the depth to the salt, the greater the overburden pressure and hence the lesser the temperature rise possible without plastic flow occurring; therefore, the size of cavity will have to be reduced to maintain the same temperature. Since the cost of the facility will be greatly dependent upon the shaft costs and the size of the storage vaults possible, it would seem that from a first cost point of view the Kansas site would be preferable to the Lake Erie Site.

There is also the possibility that for an experimental site which could not be expanded into a permanent-disposal facility, a drift mine into one of the salt outcrops in the arid west might be adequate.

It would seem that Kansas, at present, offers the greatest promise as a test site. Therefore, we have had the Geotechnical Corporation make an additional, more exhaustive report on an area in Kansas that seems to meet most of the desirable criteria for a test site.(38)

Cost of Test Facility. Assuming that a site in Kansas is chosen, the cost for setting up a test facility has been estimated by the Cost Estimating Department at Oak Ridge. These are Class "D" quotations, "based on generalized criteria, subject to further definition and change. Their degree of accuracy cannot be determined."

Costs have been estimated on two bases, temporary and permanent installations. The following assumptions were made for purposes of the estimate:

TABLE IX

COST ESTIMATE

	Temporary		Permanent	
	A	B	C	D
Shafts, diameter	84" 36"	84" 36"	84" 84"	84" 84"
Winch and cage	2 tons	2 tons	20 tons	20 tons
Filterhouse, blower, etc.	No	No	Yes	Yes
20' x 20' main tunnels, ft	500	500	1000	1000
10' x 10' side tunnels, ft	100	100	300	300
Storage pits	1 parppd*	1 sphere 1 parppd*	1 sphere 1 parppd*	1 sphere 2 parppd*
Tractor	None	None	1	1
Jeeps	1	1	2	2
Man hoist for outlet	Yes	Yes	Yes	Yes
Instrumentation	Yes	Yes	Yes	Yes
Utilities and service, etc.	Yes	Yes	Yes	Yes
	$266,000	$357,000	$529,000	$631,000

*Parallelepiped.

As shown in the cost estimate, the least amount of money for which a test site in Kansas could be prepared is $266,000. If, as is likely, more than one experiment would be carried out, then the cost will rise. There is a limit, however, to the number of experiments that can be performed on a temporary basis. If the experimental site should be constructed so that it could later be used as the permanent storage area, the initial costs will be considerably high - $529,000. If, however, the cost per gallon is considered than the incremental cost per 56,000 gals is only $102,000 or $1.82 per gallon. If the method proves successful and larger caverns can be built the price will still further decrease. Furthermore, if the method of washing cavities is successful, additional substantial reductions in the unit cost of storage will be realized.

Site Procurement. The other possible sites for the field experiment will be investigated and where suitable, a similar cost analysis will be made. The AEC will then be asked to buy or lease the land, or if Government-owned land obtain title to it for use as a field testing site.

VII. Field Experiment. After permission to use the land is obtained and the site prepared, it will be necessary to place the instrumentation in the mine. For the cold run, a synthetic waste and heaters will suffice. However, for a hot run of the order of magnitude of 2 BTU/hr/gal, over a million curies of activity will be required, assuming 93 per cent of the gamma energy and all the beta energy is effective in heating the waste, specifically 3.9×10^6 curies of cesium 137 or 3.3×10^6 curies of strontium 90.

The prospects for success in salt disposal of radioactive wastes appear to be good. The Earth Sciences Division of the National Academy of Science has stated: "Disposal in tanks is at present the safest and possibly the most economical method of storing waste. Disposal in salt is the most promising method for the near future."(8)

Our goal is an early realization of that promise.

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STATUS REPORT ON WASTE DISPOSAL IN NATURAL SALT FORMATIONS: II

by

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ABSTRACT

Radioactive liquid wastes can be stored in cavities in natural salt formations if the structural properties of salt are not adversely affected by chemical interaction, pressure, temperature, and radiation. Analytical studies show that it is possible to store two-year-old waste in a 10-foot-diameter sphere without exceeding a temperature of 200° F. Laboratory tests show that the structural properties of rock salt are not greatly altered by high radiation doses, although high temperatures increase the creep rate for both irradiated and unirradiated samples. Chemical interaction of liquid wastes with salt produces chlorine and other chlorine compound gases, but the volumes are not excessive. The migration of nuclides through the salt and deformation of the cavity can only be studied in the field in undisturbed salt in place. The design of a field experiment is complete, and the materials for construction of the cavity cover and off-gas system have been chosen.

INTRODUCTION

The first status report[1] on radioactive waste disposal in cavities in in natural salt formations characterized the limits of the problem as "the degree to which the desired structural properties of salt are affected by liquid wastes, pressure, temperature, and radiation..." This status report covers some of the theoretical considerations and preliminary laboratory investigations of these effects. The theoretical studies show that the temperature rise of radioactive wastes stored in salt cavities can be kept below acceptable limits by controlling the age of the wastes and the size of the cavity. Laboratory work on chemical compatibility shows that present-day wastes are essentially inert toward rock salt. Additionally, radiation has little effect on the structural properties of rock salt. However, all of the information necessary for the design of the field-scale radioactive experiment cannot be obtained theoretically nor in laboratory tests, because theoretical solutions require a number of simplifying assumptions, and laboratory tests of geological materials are not conclusive, because the samples are removed from their environment and, hence, disturbed. Preliminary field tests will provide information on the effects of heat and chemical interaction on the structural integrity of the cavity, the migration of nuclides from the cavity out into the surrounding salt, and the migration of the cavity itself.

THERMAL CALCULATIONS

The plasticity and structural integrity of cavities in rock salt are dependent upon temperature and pressure. Pressure is a function of the stratigraphy, the percentage of salt excavated, and the configuration of the excavation.

Temperature in a cavity is a function of its depth, size, shape, the rate of heat production of the stored material, and thermal conduction in the surrounding salt. The heat production rate, approximated from Fig. 1, is given as a function of decay time, assuming 800 gal of waste per metric ton of uranium, irradiated for 10,000 Mwd/ton at 33 Mw/ton specific power.[2]

If a limit to the temperature rise within a cavity and the space about it is predetermined, then the time-space-temperature distribution for various sizes, shapes, and heat production rates of the wastes stored within the cavity can be calculated.

Birch has stated "that heating the liquid some hundred degrees (centigrade) or so would not produce drastic physical changes, and that plastic deformation of shale or evaporite layers, if sufficiently thick might continue to provide a tight container."[3] We, therefore, set a 200° F rise as the maximum permissible temperature rise in a salt cavity.

The possible sizes and shapes of cavities and the heat production rates of wastes are so numerous that a few simplifying assumptions were necessary:

(1) Only spherical shapes would be investigated so that an analytical solution would be possible.

(2) Diameters of the spheres would be limited to 10 ft and 20 ft, since it had been shown previously[4] that larger spheres, containing wastes generating heat at a rate of 2 BTU/hr/gal, would exceed the permissible limit of 200° F.

(3) Wastes considered would be limited to those described by Fig. 1 and cooled for 1, 6, and 11 years.

(4) Waste-filled cavities would be located in an infinite body of salt.

(5) Thermal conductivity, density, and specific heat of the salt and the wastes would be constant and independent of changes in temperature.

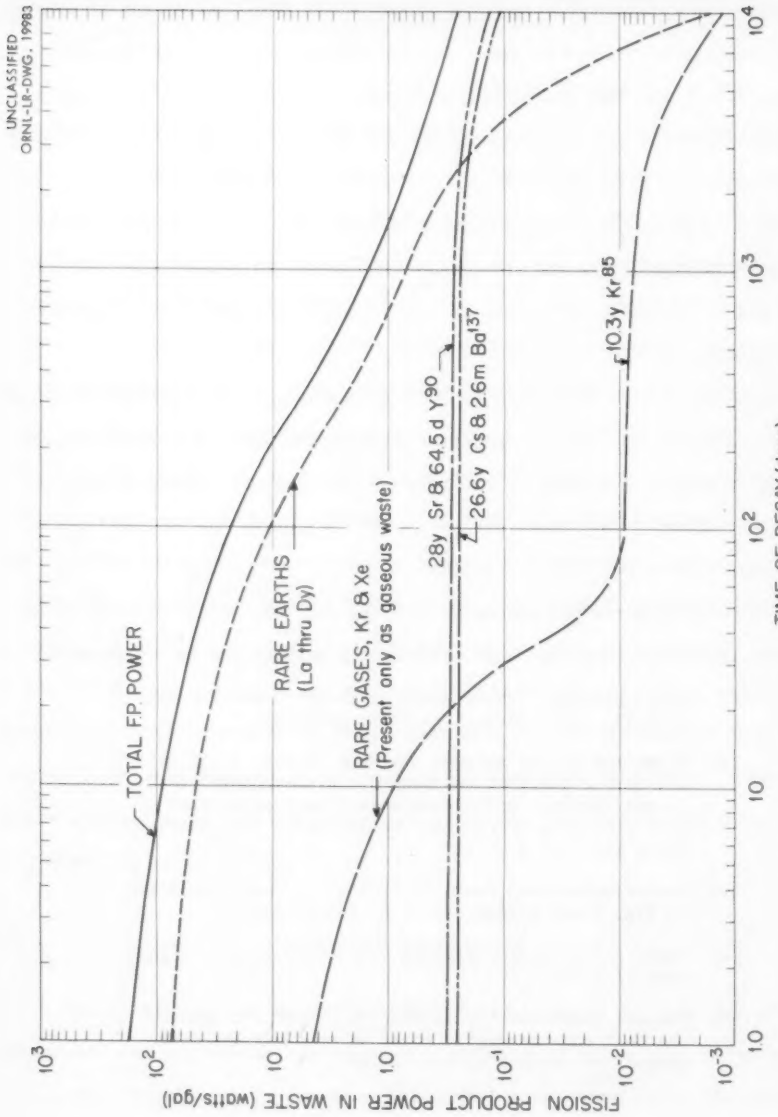

FIG. 1. FISSION PRODUCT POWER IN WASTE AS A FUNCTION OF DECAY TIME BASIS: 800 GAL WASTE PER METRIC TON U, IRRADIATED 10,000 Mwd/ton At 33 Mw/ton SPECIFIC POWER.

(6) Heat would be lost by conduction only.

(7) Phase changes would not occur in the wastes or in the salt.

(8) Convective currents would not develop in the wastes.

(9) All beta and gamma energy would be absorbed in the wastes.

All assumptions, except the extent of salt, are on the safe side; that is, the temperatures calculated would be higher than those actually achieved. The absence of convective currents in the wastes would produce a temperature gradient from the center of the cavity out to the interface of the salt and the waste. If convective currents were present, the temperature would be uniform throughout the sphere. For cavities of the size considered, however, the edge temperature in the conduction case would approach the uniform temperature achieved if convective currents were present.

Statement of Problem

A mathematical statement of the spherical problem is:

$$\frac{1}{\alpha_1}\frac{\partial T_1}{\partial t} = \frac{\partial^2 T_1}{\partial r^2} + \frac{2}{r}\frac{\partial T_1}{\partial r} + \frac{Q(t)}{K_1}, \qquad 0 \leq r < a,\ t > 0, \quad (1)$$

and

$$\frac{1}{\alpha_2}\frac{\partial T_2}{\partial t} = \frac{\partial^2 T_2}{\partial r^2} + \frac{2}{r}\frac{\partial T_2}{\partial r}, \qquad r > a,\ t > 0, \quad (2)$$

where subscripts 1 and 2 refer to the heated sphere and to the infinite body, respectively; T is temperature; t is time; r is radial distance from the center

PER METRIC TON U, IRRADIATED 10,000 Mwd/ton At 33 Mw/ton SPECIFIC POWER.

of the sphere; a is the radius of the sphere; K is thermal conductivity; α is thermal diffusivity; and Q(t) is heat generation per unit time per unit volume.

Initially, the wastes and salt are considered to be at zero temperature. Boundary conditions are such that the temperature and heat flux are continuous at the interface, and the temperature remains finite both at the center of the sphere and at infinity. Then, by the use of La Place Transforms and Duhamel's theorem the following equations were obtained. The temperature inside the cavity is

$$T_1(t) = \int_0^t \sum_{i=1}^{N} A_i \frac{e^{-\lambda_i(t-\tau)}}{2C_2} \left\{ \operatorname{erf}\left[\frac{a-r}{2\sqrt{\alpha_2\tau}} \right] + \operatorname{erf}\left[\frac{a+r}{2\sqrt{\alpha_2\tau}} \right] - \sqrt{\frac{4\alpha_2\tau}{r^2\pi}} \left[e^{-(a-r)^2/4\alpha_2\tau} - e^{-(a+r)^2/4\alpha_2\tau} \right] \right\} d\tau,$$

and outside of the cavity

$$T_2(t) = \int_0^t \sum_{i=1}^{N} A_i \frac{e^{-\lambda_i(t-\tau)}}{2C_2} \left\{ \operatorname{erf}\left[\frac{r+a}{2\sqrt{\alpha_2\tau}} \right] - \operatorname{erf}\left[\frac{r-a}{2\sqrt{\alpha_2\tau}} \right] - \sqrt{\frac{4\alpha_2\tau}{r^2\pi}} \left[e^{-(r+a)^2/4\alpha_2\tau} - e^{-(r-a)^2/4\alpha_2\tau} \right] \right\} d\tau,$$

where N is the number of fission products. A_i is the heat generation per unit time per unit volume for ith fission product, and λ_i is the disintegration constant of the ith fission product as $\sum_{i=1}^{N} A_i e^{-\lambda_i t}$ is substituted for Q(t). C is the specific heat per unit volume ($C = \frac{K}{\alpha}$); τ is a variable of integration; and erf is the error

function. For rock salt, K_2 equals 2.07 $\frac{\text{BTU}}{\text{ft-hr-}^\circ\text{F}}$; α equals 0.07 $\frac{\text{ft}^3}{\text{hr}}$; and C_2 equals 29.7 $\frac{\text{BTU}}{\text{ft}^3\text{-}^\circ\text{F}}$.

Summing the integrals of the decay curves of all fission products contributing to heat generation would be arduous. Therefore, the total fission product power, as shown on Fig. 1, is approximated by six exponentials: The decay curves for Cs^{137} and Ba^{137}, Sr^{90} and Y^{90}, long-lived components of the rare earths, short-lived components of the rare earths, long-lived components, and short-lived components of the remainder of the fission products.

Results

Equations for the temperatures both inside and outside the cavity were numerically integrated on the IBM 704 using Simpson's Rule and the Fortran coding system to calculate the temperature rises.[5] Curves of these data are shown in Figs. 2 to 7.

For the 10-foot-diameter sphere and one-year-old waste (Fig. 2), it is seen that the maximum rise in temperature at the center of the sphere is 530° F at 1000 hr and 315° F at the surface of the sphere at 1500 hr. In Fig. 3 the maximum temperature rise for six-year-old waste at the center of the sphere is 100° F at 4500 hr and 65° at the surface at 7000 hr. In Fig. 4 the maximum temperature rise at the center of the sphere of eleven-year-old waste is 65° F at 7000 hr and 40° F at the surface at 10,000 hr. As interpolated from these data, two-year-old wastes stored in a 10-foot-diameter sphere would reach a peak temperature of 200° F in less than five months.

For the 20-foot-diameter sphere (Figs. 5, 6, and 7), it is seen that the same wastes could not be stored in cavities of this size without boiling.

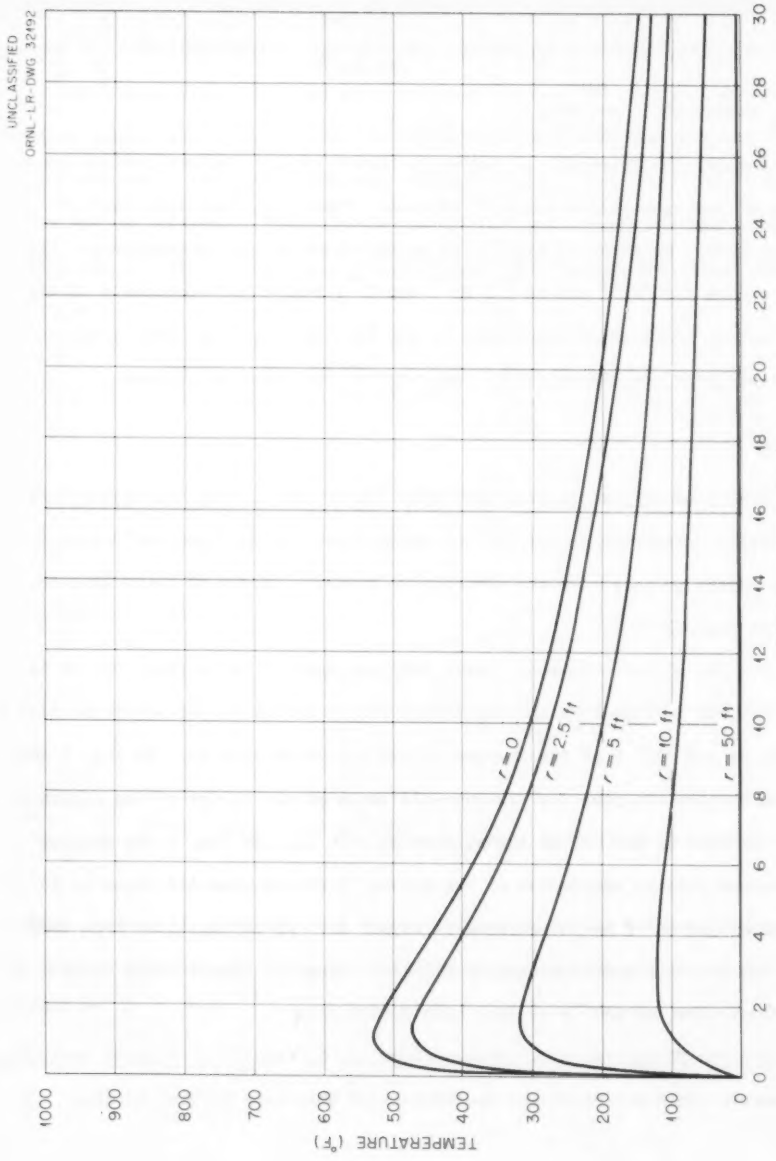

Fig. 2. 1 Year Old Waste – Sphere of 5-ft Radius, Surrounded by an Infinite Bed of Salt.

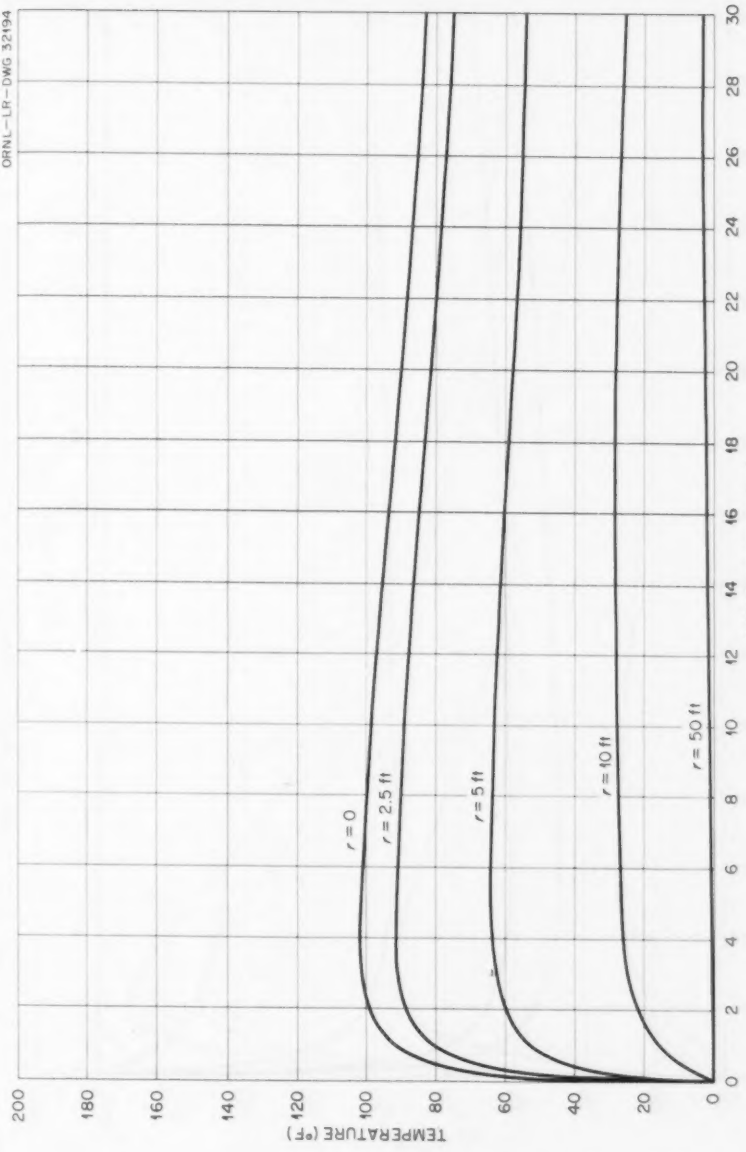

Fig. 3. 6 Year Old Waste – Sphere of 5-ft Radius. Surrounded by an Infinite Bed of Salt.

Fig. 2. 1 Year Old Waste – Sphere of 5-ft Radius, Surrounded by an Infinite Bed of Salt.

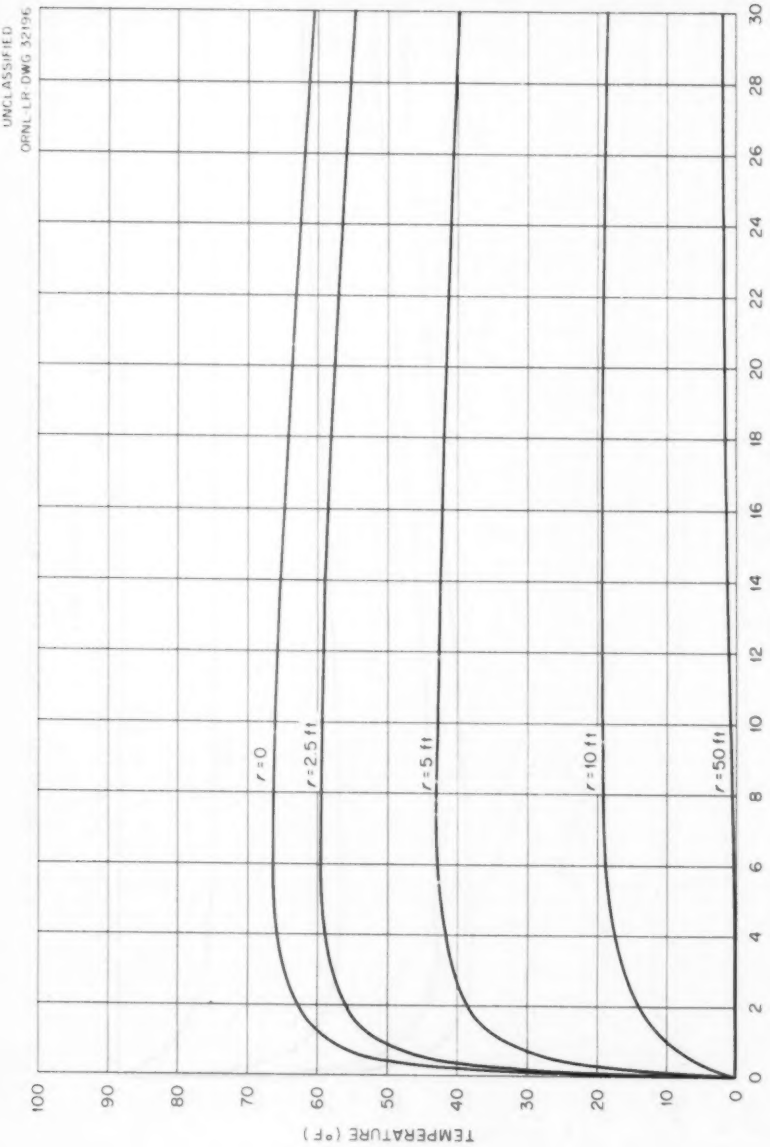

Fig. 4 11 Year Old Waste–Sphere of 5-ft Radius. Surrounded by an Infinite Bed of Salt

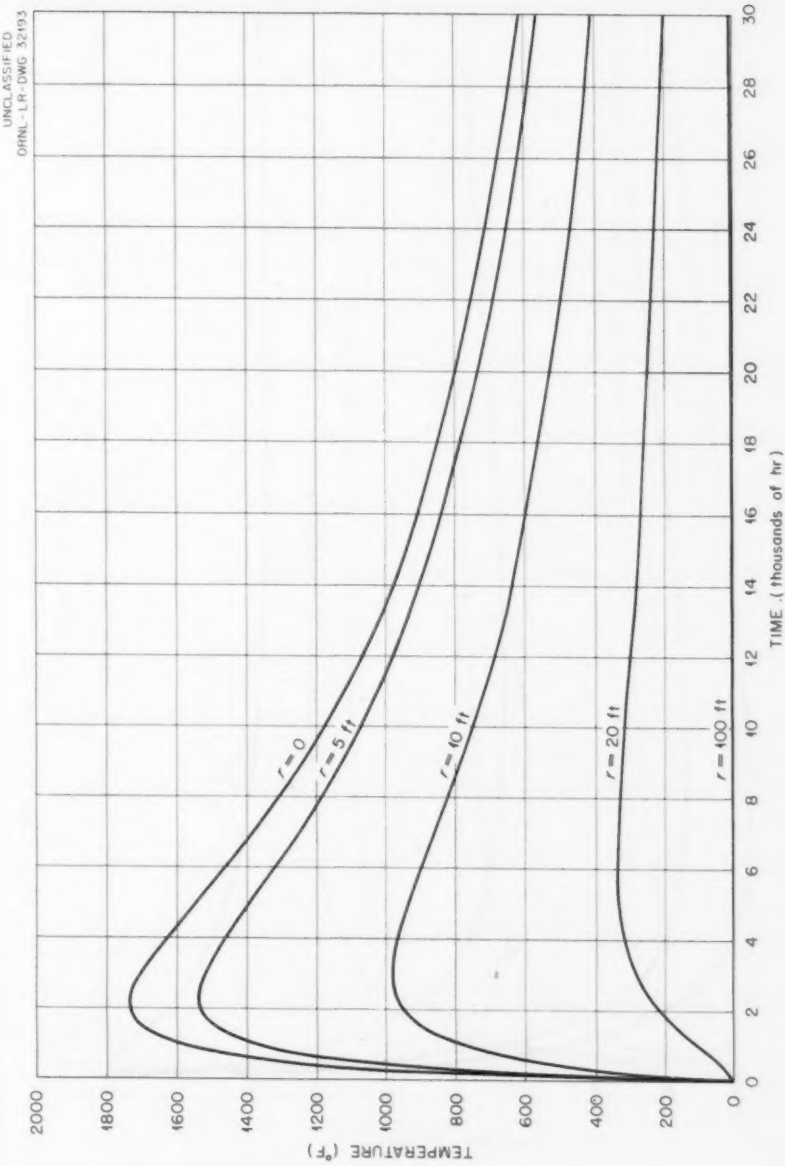

Fig. 5. 1 Year Old Waste – Sphere of 10-ft Radius, Surrounded by an Infinite Bed of Salt.

Fig. 4. 1 Year Old Waste-Sphere of 5-ft Radius, Surrounded by an Infinite Bed of Salt

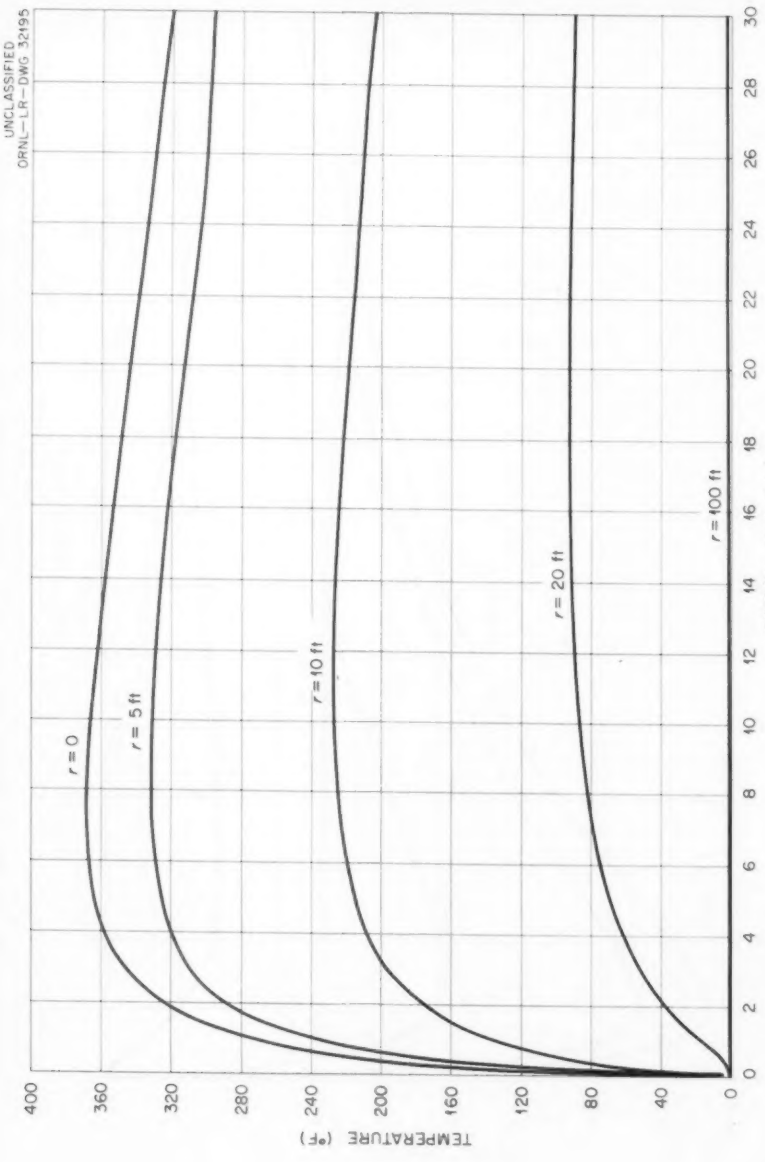

Fig. 6. 6 Year Old Waste–Sphere of 10-ft Radius. Surrounded by an Infinite Bed of Salt.

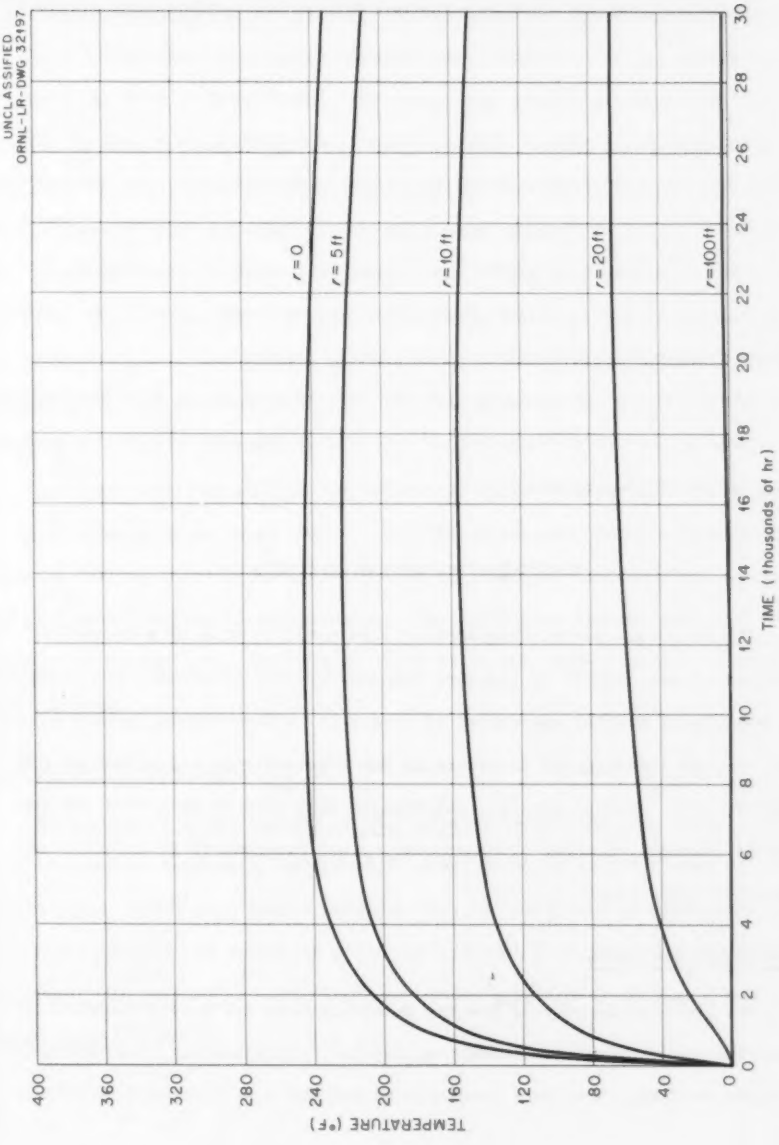

Fig. 7. 11 Year Old Waste – Sphere of 10 – ft Radius. Surrounded by an Infinite Bed of Salt.

These results are substantially the same as the calculations of Birch[6] and Schecter.[7] They considered only temperature-time distributions for the conduction case; in addition, they considered the convection case in their calculations. For a sphere of 5-foot radius, Schecter found a drop of 35% from center-of-the-sphere temperature in the conduction case to uniform sphere temperature in the convection case. Our calculations for spheres of this size show a 40% reduction from center temperature to surface temperature in the conduction case, or little difference between surface temperature in the conduction case and sphere temperature in the convection case.

Therefore, we may conclude that the thermal problem in salt disposal will cause no serious difficulties if the size of the cavities and the age and dilution of the wastes are controlled.

STRUCTURAL PROPERTIES OF ROCK SALT

The rock salt surrounding waste-filled cavities will be subjected to certain stresses induced by elevated temperatures and pressures, and radiation. The response of crystal aggregates of rock salt to temperature, pressure, and radiation was investigated to determine the stress-strain relationships and the creep rate.[8] Stress-strain relationships were used to determine the compressive strength, modulus of elasticity, yield strength (0.2% offset), and apparent elastic limit.

Experimental Procedure

The American Society of Testing Materials does not have a standard procedure for the testing of rock-salt aggregates. Consequently, the test procedures for wood and stone were investigated and the more pertinent provisions

applied to the testing of rock salt.[9] Two-inch cubes of salt, randomly selected, were used as test specimens. Specimens of dome salt were machined from a large block taken from the Morton Salt Company's mine at Grand Saline, Texas. Specimens of bedded-deposit salt were machined from a block taken from the Carey Salt Company's mine at Hutchinson, Kansas. The purity of dome salt was greater than 98% NaCl and that of the bedded salt was 95% NaCl.

Machined specimens were irradiated in the Cobalt Storage Facility of the Operations Division at ORNL. Approximately 200,000 curies of Co^{60} in aluminum clad cylinders are stored about a one cubic foot chamber below ground level to produce an exposure dose rate of 3.2×10^6 roentgens per hour, as measured with a ceric sulfate chemical dosimeter.

Specimens were tested with the assistance of the Mechanical Testing Section of the Metallurgy Division at ORNL. A 120,000-lb capacity Baldwin-Tate-Emery Universal testing machine was used for load application. Deformation was measured by a Bell-Crank-Anvil extensometer. This apparatus automatically plotted a total load versus total deformation curve which was then reduced to a stress-strain diagram.

Tests at Room Temperature

Stress-Strain Relationships of Unirradiated Bedded and Dome Salt.-- Three specimens each of dome salt, bedded salt loaded parallel to the planes of stratification, and bedded salt loaded perpendicular to the planes of stratification were tested in order to establish a control with which to compare irradiated specimens. The results of these tests are shown graphically in Fig. 8 and in tabular form in Table 1.

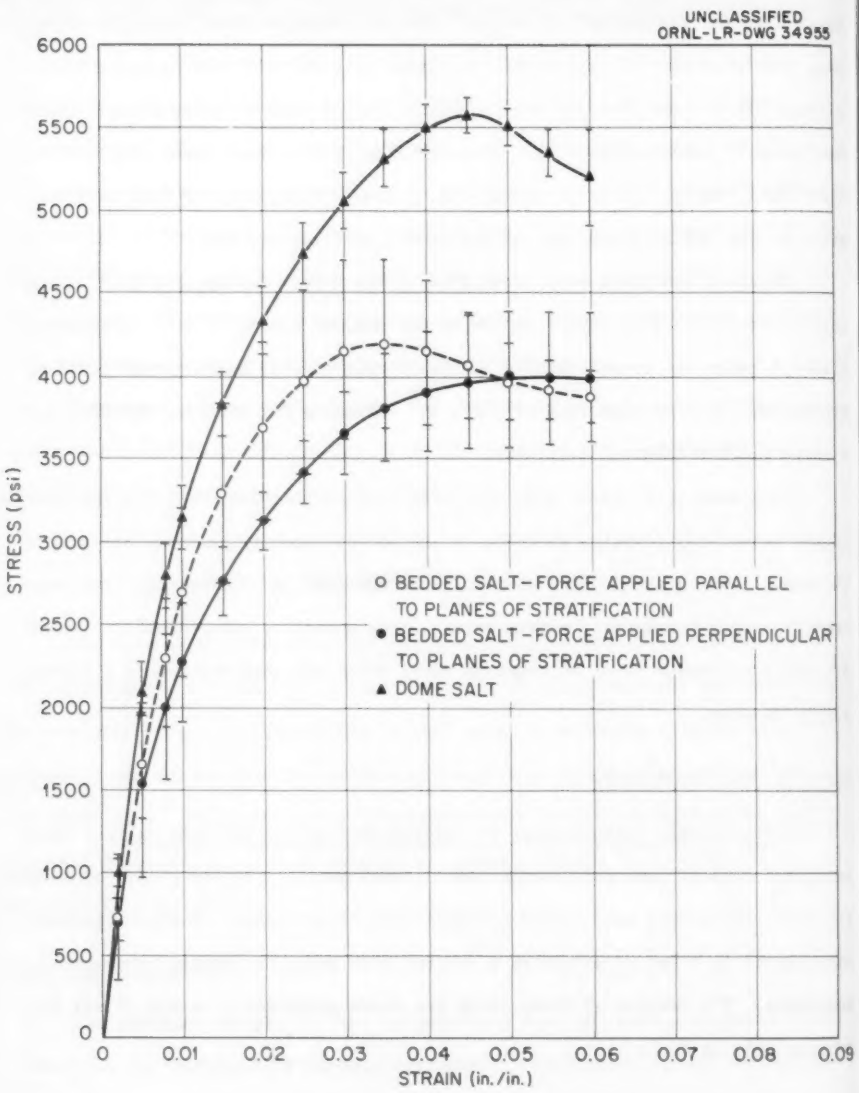

Fig. 8. Compression Test of Unirradiated Salt at Room Temperature.

Table 1. Structural Properties of Rock Salt

Type of Salt	Temperature	Exposure Dose(r)	Compressive Strength (psi)	Yield Strength (psi)	Apparent Elastic Limit (psi)	Modulus of Elasticity (psi) x 10^6
Dome Bedded Parallel*	Room	0	5600 ± 100[a]	2900 ± 150	2000 ± 100	.50 ± .05
Bedded Perpendicular**	Room	0	4200 ± 500	2500 ± 300	1900 ± 300	.35 ± .06
Bedded Parallel	Room	0	4000 ± 400	2300 ± 300	1700 ± 200	.36 ± .17
Bedded Parallel	Room	10^6	4200 ± 500	2500 ± 300	1900 ± 300	.35 ± .06
Bedded Parallel	Room	10^7	4500 ± 500	2500 ± 150	1900 ± 200	.32 ± .06
Bedded Parallel	Room	10^8	4600 ± 400	2700 ± 250	2100 ± 200	.35 ± .02
Bedded Parallel	Room	5×10^8	4600 ± 150	3400 ± 250	2800 ± 200	.36 ± .09
Bedded Parallel	200° C	0	3500 ± 50	2700 ± 200	2000 ± 300	.39 ± .07
Bedded Parallel	200° C	5×10^8	3900 ± 200	1600 ± 200	1400 ± 100	.090 ± .01
			3400 ± 400	1900 ± 250	1800 ± 250	.12 ± .02

* Bedded parallel - bedded salt, force applied parallel to the planes of stratification.
** Bedded perpendicular - bedded salt, force applied perpendicular to the planes of stratification.
a - All results have been rounded off to the nearest 100 psi and the standard deviation to the nearest 50 psi.

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Dome salt has greater compressive and yield strengths than bedded salt. Previous work with synthetic crystals indicated that small amounts of impurities added to sodium chloride increased the tensile strength of salt. The results for synthetic and single crystals are not applicable to aggregates of crystals.[10] The binding strength between crystals becomes an important factor in the strength of the aggregates. The presence of other substances in the specimens, anhydrites and shales, is at least partially responsible for the greater variation exhibited by the bedded salt. Dome salt, although neither isotropic nor homogeneous, is much more uniform with respect to impurities than bedded salt. The mined block of bedded salt was interstratified with anhydrite and shale bands of various thicknesses.

Bedded salt is slightly stronger when loaded parallel to planes of stratification than when loaded perpendicularly. Although this difference is within the statistical variation of the experiment, the same trend is evident in creep tests.

Creep Tests - Unirradiated Bedded and Dome Salt.-- The creep rate resulting from the application of a constant load of 2500 pounds per square inch for 100 minutes was measured. The creep rates of three types of salt are shown in Fig. 9. After an initial rise, due mostly to elastic compression, the strain increases very slowly with time. As in the stress-strain tests, dome salt is strongest (least creep under the same load conditions) and exhibits the least variation from sample to sample. Bedded salt loaded parallel to the planes of stratification is again stronger than bedded salt load perpendicular to the planes of stratification, though still within the statistical variation of the experiment.

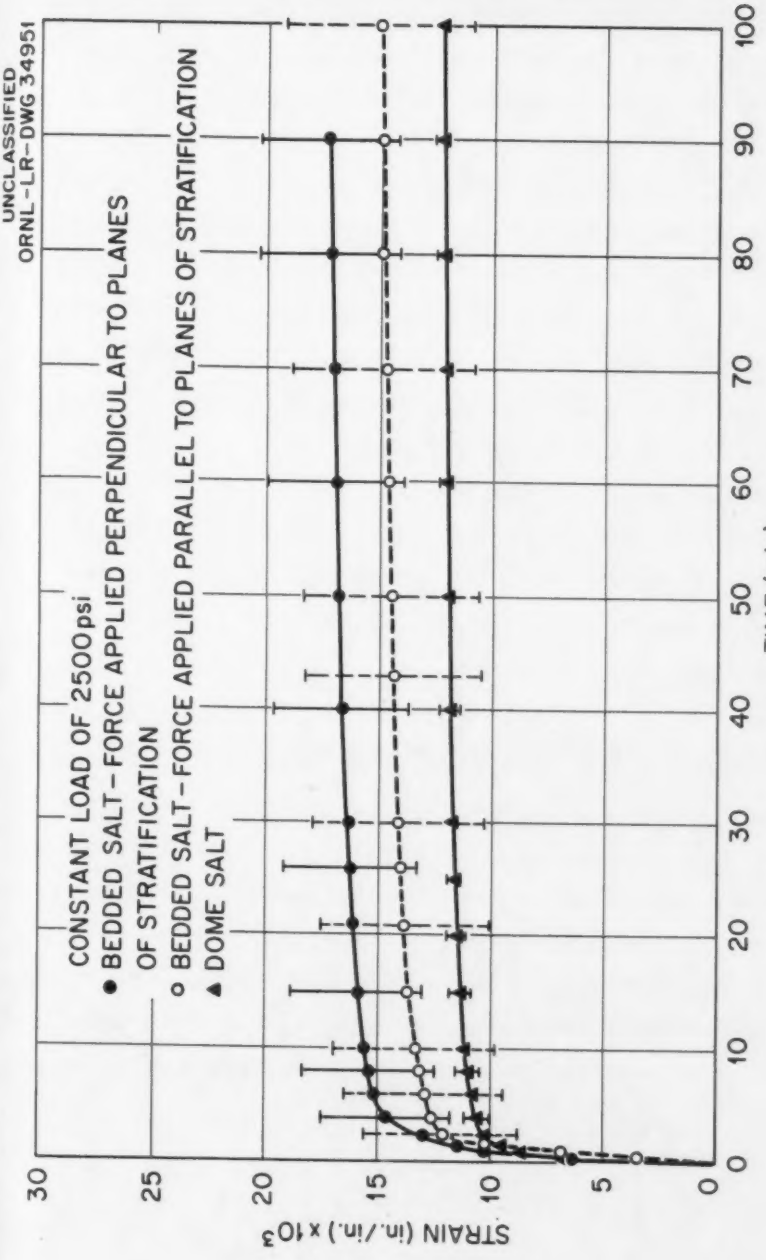

Fig. 9. Creep Test of Unirradiated Salt at Room Temperature.

Stress-Strain Relationship of Irradiated Bedded Salt Loaded Parallel to the Planes of Stratification.-- Bedded salt samples were irradiated for periods to give exposure doses of 10^6, 10^7, 10^8, and 5×10^8 roentgens, assuming a dose rate constant with time.

The stress-strain curves of irradiated bedded salt are shown in Fig. 10, and the data are tabulated in Table 1. From Fig. 10 it is seen that there is very little difference in the stress-strain relationship up to a stress of 2000 pounds per square inch. The compressive strength increases with exposures up to 5×10^8 r, where a marked decrease is observed. The compressive strengths up to exposures of 5×10^8 r are within the statistical variation of the experiment, while the low value obtained at 5×10^8 is not.

The expected increase in the modulus of elasticity is observed only at 5×10^8 r and is within the limit of statistical variation. In fact, at 10^6 r the modulus of elasticity, yield strength, and apparent elastic limit are slightly lower than for the unirradiated specimens.

The yield strength and apparent elastic limit reach a maximum at 10^8 r. However, at 5×10^8 r the yield strength and apparent elastic limit, unlike the compressive strength, are greater than for the unirradiated specimens.

Creep Rate of Irradiated Bedded Salt Loaded Parallel to the Planes of Stratification.-- The effect of radiation on the rate of creep was investigated by testing three specimens given 5×10^8 r. The results were compared with the results for unirradiated salt (Fig. 11).

Very little difference in the creep rate can be seen between the irradiated and unirradiated salt. Only a little over a thousandth of an inch strain separates the two curves. The unirradiated salt actually shows less strain than

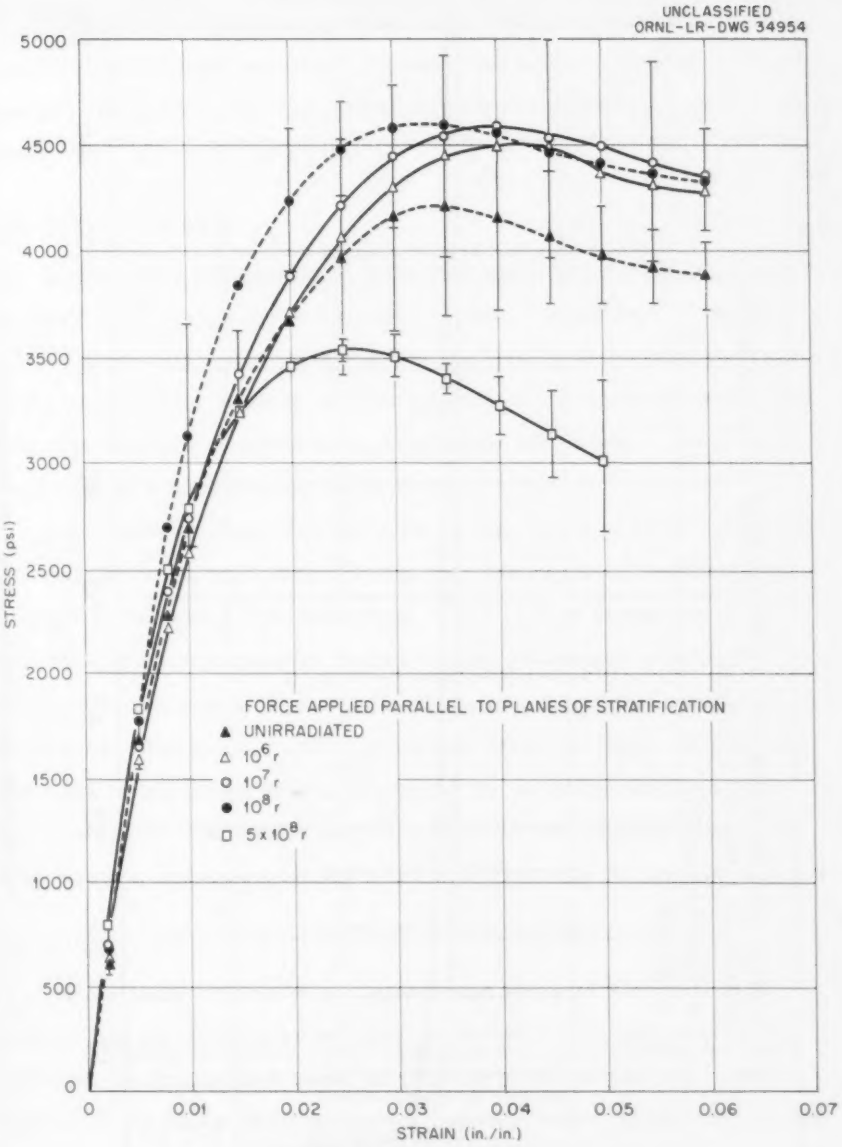

Fig. 10. Compression Test of Bedded Salt at Room Temperature.

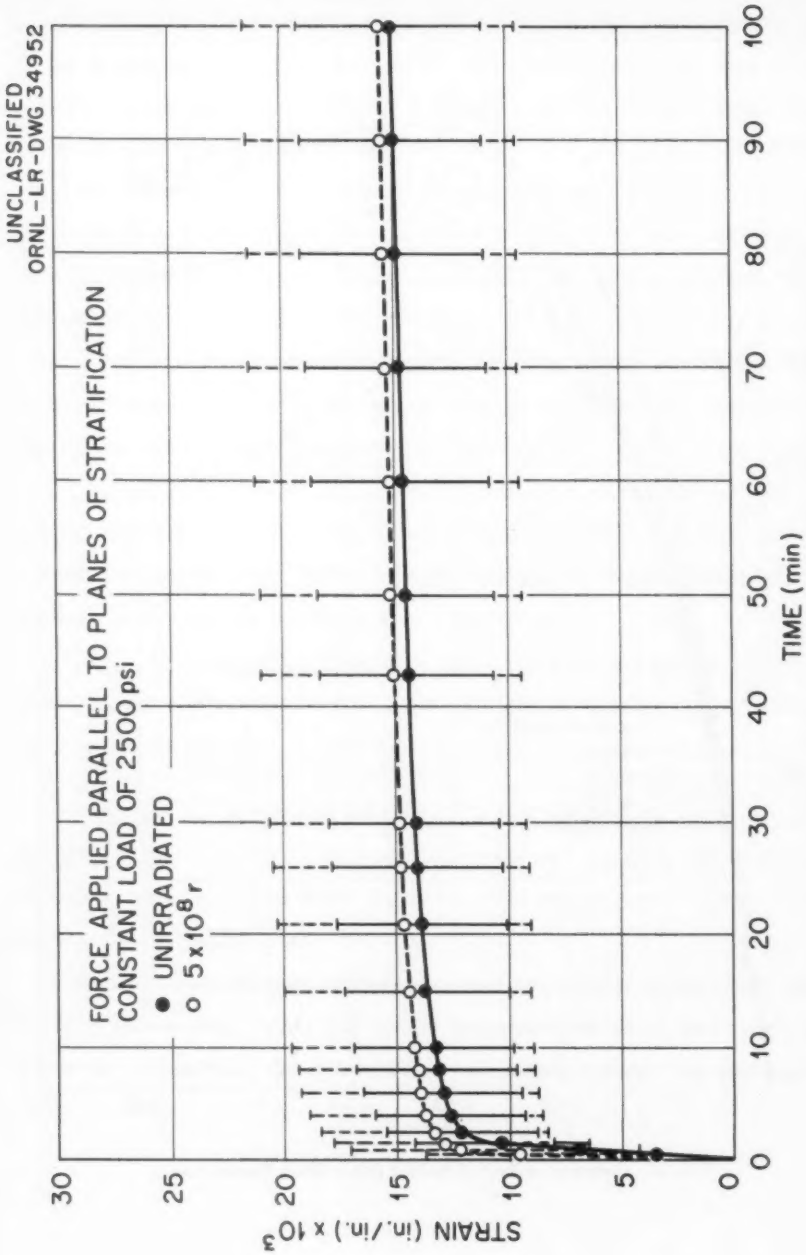

Fig. 11. Creep Test of Bedded Salt at Room Temperature.

the irradiated specimen. If the modulus of elasticity is increased, the irradiated salt should show less strain. However, two of the irradiated specimens developed cracks during the test, resulting in a wide variation in results and probably causing the greater strain.

Tests at 200° Centigrade

Stress-Strain Relationship of Irradiated Bedded Salt Loaded Parallel to the Planes of Stratification.-- The expected rise in temperature of the salt surrounding waste-filled cavities suggests the need for determining the structural properties of irradiated and nonirradiated salt at elevated temperatures. Three specimens each of bedded salt, unirradiated and exposed to 5×10^8 r, were tested at a temperature of 200° C.

The results of these tests are given in Fig. 12 and in Table 1 (note that the strain scale has been changed in Fig. 12). As expected, the plasticity increases quite rapidly with temperature, with a maximum deformation of 25%. Also, the modulus of elasticity, yield strength, and apparent elastic limit are lower than at room temperature. As in the room temperature tests, the stress-strain response at 5×10^8 r rises much faster initially and falls off more rapidly than the stress-strain response for the unirradiated specimens. This gives, as at room temperature, an increase in the modulus of elasticity, yield strength, and apparent elastic limit and a decrease in compressive strength.

Fig. 11. Creep Test of Bedded Salt at Room Temperature.

Conclusions

On the basis of these tests, unirradiated dome salt is consistently stronger than unirradiated bedded salt. Under both static and creep loading conditions the unirradiated bedded salt appears to be stronger when loaded parallel to the planes of stratification than when loaded perpendicular to the

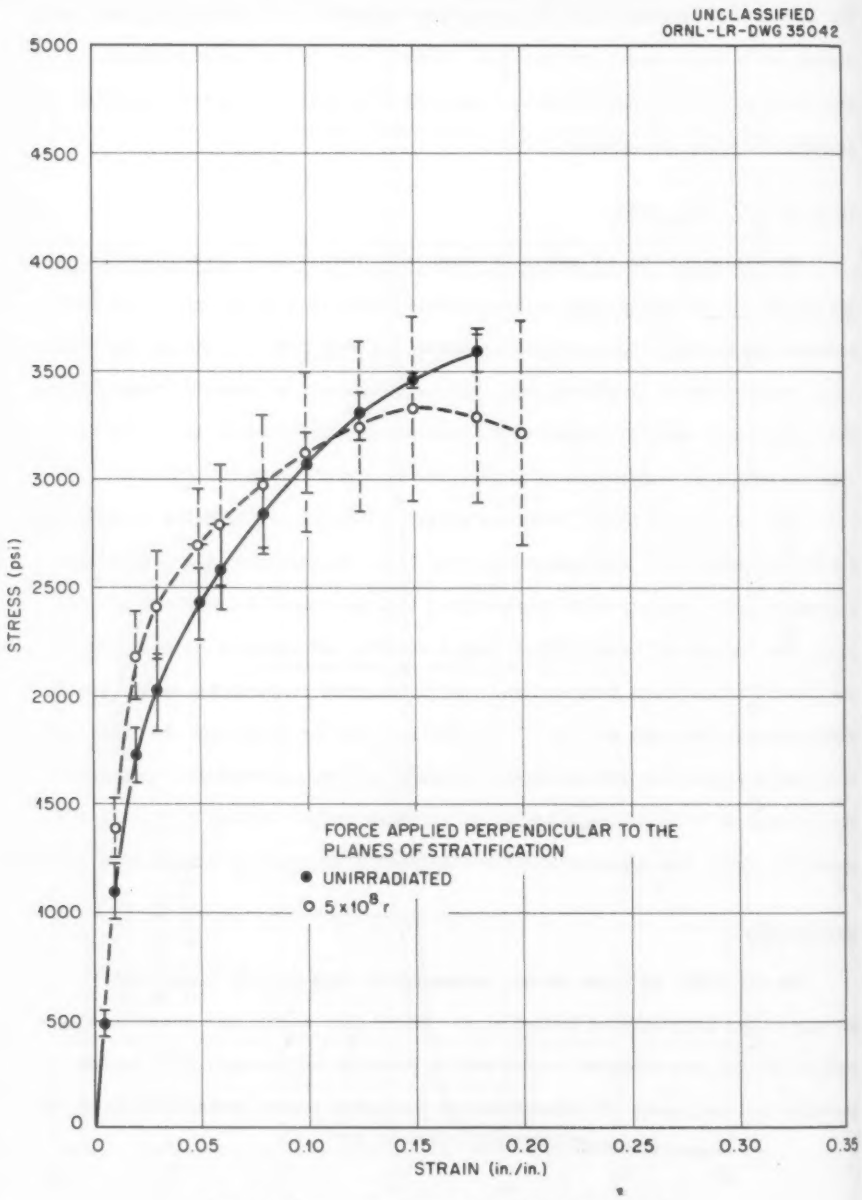

Fig. 12. Compression Tests of Bedded Salt at 200°C.

planes of stratification. However, the difference is within the statistical variation of the tests. Radiation causes only minor changes, if any, in the structural properties of salt. The most significant changes are: (1) a decrease in the compressive strength at an exposure dose of 5×10^8 r, (2) an increase in the modulus of elasticity at an exposure dose of 5×10^8 r, and (3) an increase in the yield strength and apparent elastic limit at all exposure doses except 10^6 r. However, with one exception (the compressive strength at 5×10^8 r), all are within the statistical variation of the tests. It appears that the effects of radiation on the structural properties of rock salt are minor.

CHEMICAL INTERACTION OF WASTE AND SALT

The safe disposal of radioactive wastes in rock salt is dependent in part upon the chemical interaction between the wastes and the salt. In view of the large number of variables and possible reactions in acid or neutralized waste-salt mixtures, it was felt that oxidation-reduction reactions should be investigated initially. The study of radiation-induced reactions would come later.

Unirradiated Mixtures

Nitric acid in the Purex waste and sodium chloride in the salt constitute the bulk of available reactants; however, other materials must also be considered. For example, natural salt includes associated deposits of mineral carbonates, sulfates, and other chloride salts, while the wastes contain small quantities of corrosion products and organic complexing agents.

Samples of salt were selected from the Kleer Salt Mine (Grand Saline, Texas), the International Salt Mine (Detroit, Michigan), and the Carey Salt Mine

(Hutchinson, Kansas). The Texas salt, typical of dome salt, was very pure (98%), whereas the other two samples of bedded salt ranged in purity from 87% to 94%. Synthetic, nonradioactive Purex waste solution was prepared according to ORNL CF-58-4-45.[11]

Preliminary experiments indicated that precipitation and thermal reactions between synthetic Purex waste and salt were of little importance. Gas production was found to be the most reliable index of chemical interaction. To study the effect of acidity, temperature, and salt purity on gas production, a series of acidified and neutralized waste-salt mixtures were prepared and stored in a temperature controlled (± 1° C) water bath. The total pressure and volume of gas produced by these mixtures at 25°, 60°, and 80° C, were measured with a gas burrette and water manometer, and collected for quantitative analysis. The results of these tests at 25° C are shown in Fig. 13. The gas produced in the Texas salt-waste mixture occurs immediately, whereas the Kansas and Michigan salt-waste mixtures produce larger volumes of gas at a much slower rate. Neutralized waste-salt mixtures produce gas at about the same rate as the control samples. These tests indicate that the system behaves according to the usual oxidation-reduction reactions, i.e., the rate of reaction increases with increases in concentration and temperature.

Quantitative and qualitative analyses of the gas showed that NO_2 was produced in trace amounts in the neutralized waste-salt mixture and in small amounts (less than 2% by volume) in the acid waste-salt mixtures. The major constituent in the gas from acid waste-salt mixtures was CO_2. If Cl_2 and NO_2Cl were produced, the concentration was below the detectable limits of the ORSAT gas analysis equipment used in these tests.

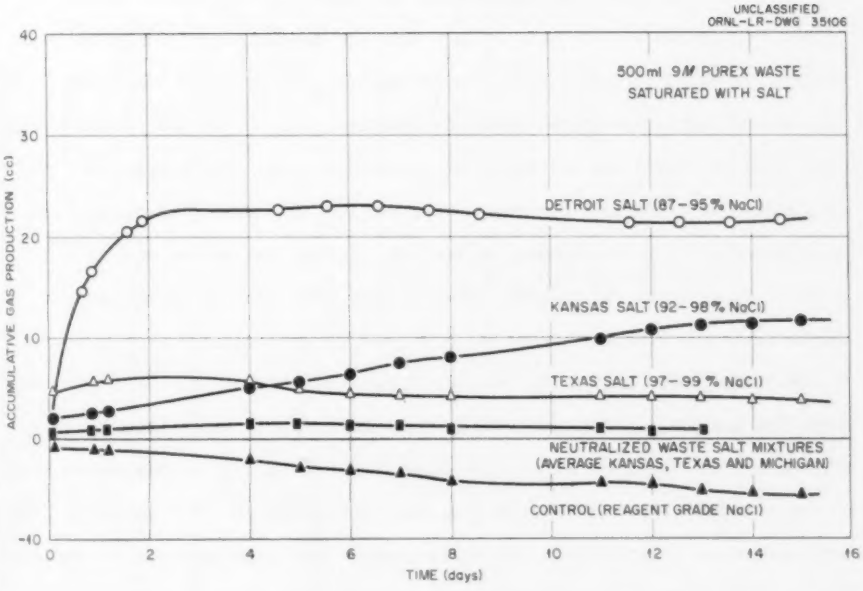

Fig. 13. Gas Production of Unirradiated Purex Type Waste.

Irradiated Mixtures

The effect of absorbed beta and gamma radiation on these reactions can best be studied by placing radioactive wastes in rock salt. However, some information can be obtained from waste-salt mixtures exposed to Co^{60} gamma radiation.

Waste-salt mixtures were placed in duplicate one-liter Pyrex bottles equipped with vent tubes. The bottles were placed in the Cobalt Storage Facility and connected to the gas measurement apparatus with 1/8-inch I. D. plastic tubing through the thermocouple wells. This arrangement permitted volume measurement and sampling during the radiation period. The recorded temperature was the average temperature of the radiation chamber. All of the gas produced during the radiation period was collected and analyzed with the ORSAT apparatus. The cumulative gas production (average of two samples) for 500-ml waste-salt mixtures exposed for 30 hr is shown in Table 2. Neutralized wastes with no salt (Run No. 4) produced the greatest volume of gas (1120 cc), while neutralized wastes saturated with salt (Runs No. 5, 8, and 10) produced the next largest volumes, 1090, 925, and 900 cc, respectively. The smallest volumes of gas, between 300 and 600 cc, were produced by the saturated acid waste (Runs 3, 7, 9, 11, and 13) and the unsaturated acid wastes (Runs 6 and 15). A cumulative record of each of these types is shown in Fig. 14. Gas production from irradiated neutralized wastes is about 90 to 100 times greater than from unirradiated wastes, and gas production from irradiated acid waste is about 15 to 30 times greater than from unirradiated waste. The difference in gas production between the saturated and unsaturated salt-waste mixtures is proportional to the differences in vapor pressure for the two mixtures (Runs 4 and 10). Qualitative analysis

Table 2. Cumulative Gas Production of Waste and Waste-Salt Mixtures Exposed to Gamma Radiation

Radiation field, 3.5×10^6 r/hr
Initial Sample Volume, 500 ml

Run	Description of Solution	Saturated with Salt	Cumulative Gas Production (cc)				
			Irradiation Time				
			5.0 hr	10.0 hr	15.0 hr	20.0 hr	30.0 hr
3A	8 M Purex, acid	Yes	143.7	220.7	328.3	420.9	562.0
4A	8 M Purex, adjusted to pH 10.0	No	275.4	443.6	622.1	811.9	1116.8
5A	8 M Purex, adjusted to pH 10.0	Yes	355.9	555.8	686.1	798.3	1086.2
6A	8 M Purex, acid	No	248.4	279.1	295.0	320.4	382.8
7A	4 M Purex, acid	Yes	221.7	230.3	234.8	248.9	
8A	4 M Purex, adjusted to pH 10.0	Yes	290.4	430.7	549.2	689.8	925.2
9A	1.88 M Purex, acid	Yes	215.6	239.9	279.1	317.1	344.2
10A	8 M Purex, adjusted to pH 10.0	Yes	264.1	420.9	547.9	651.1	902.2
11A	5 M Purex, acid	Yes	263.6	300.0	311.0	319.4	396.7
12A	6.27 M Purex, acid	Yes					
13A	8 M Purex, acid	Yes	291.6	349.8	360.6	381.0	479.0
14A	Salt slurry, 8 M Purex, acid	Yes	459.5	505.3			
15	8 M Purex, acid	No	292.8	322.1	339.3	402.7	506.0

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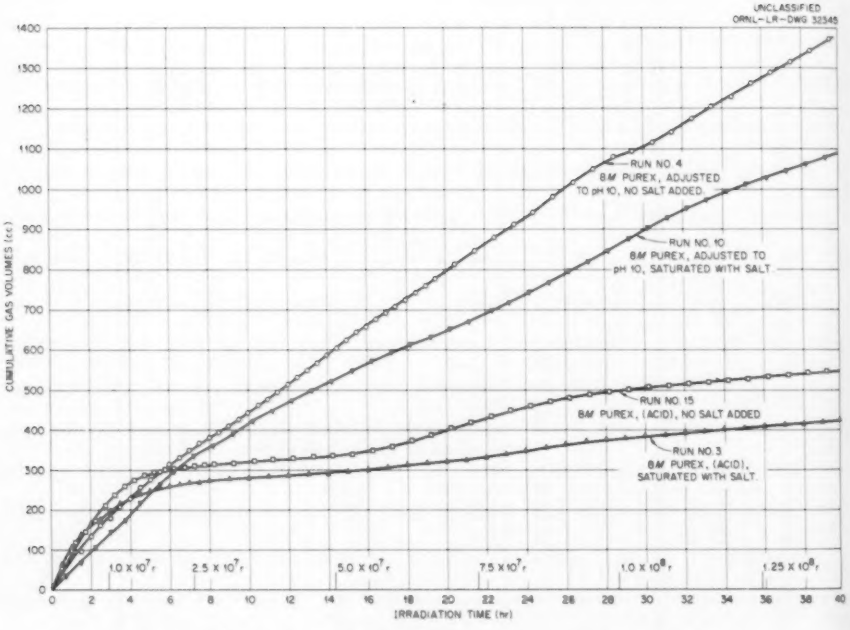
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CUMULATIVE GAS VOLUMES (cc)
IRRADIATION TIME (hr)
RUN NO. 4
8M PUREX, ADJUSTED TO pH 10, NO SALT ADDED.
RUN NO. 10
8M PUREX, ADJUSTED TO pH 10, SATURATED WITH SALT.
RUN NO. 15
8M PUREX, (ACID), NO SALT ADDED
RUN NO. 3
8M PUREX, (ACID), SATURATED WITH SALT.
1.0 X 10^7 r
2.5 X 10^7 r
5.0 X 10^7 r
7.5 X 10^7 r
1.0 X 10^8 r
1.25 X 10^8 r

Fig. 14. Gas Production in Purex-Type Waste Subjected to Gamma Irradiation.

of the gases produced in acidic and basic solutions (Tables 3 and 4) shows very little difference in composition; however, there is an increase in hydrogen and oxygen concentrations in the neutralized solution.

These results indicate that gases produced in acidified and neutralized waste-salt mixtures are qualitatively similar and probably derived from the same reactions. The increased hydrogen and oxygen concentrations in neutralized solutions may be the result of an additional reaction or the increased radiolysis of water. One physical difference in the two mixtures, which seems to correlate with increased gas production, is the presence of a hydrous iron oxide precipitate in the neutralized solutions. This precipitate increases the concentration of solid reactants available for gamma ray interaction which may increase the number of secondary electrons available for chemical interaction.

Model of the Field Test

Attempts were made to use small samples of salt (1- and 2-inch cubes) as experimental storage containers for acid and neutralized waste solutions. Invariably, however, these small samples of mined salt were found to have cracks or fractures and could not be used. Larger salt blocks (~ 2 ft on a side), as shown in Fig. 15, with hand-excavated cavities approximately 7 1/2 inches square and 9 1/2 inches deep were used instead. A Teflon cover, reflux condenser, 200-watt quartz-sheathed immersion heater, and thermocouples located in the cavity, as well as in the salt block, (locations given in Table 5) were used to test a small-scale version of a field experiment. Six liters of 7 molar, neutralized, synthetic Purex waste, saturated with the salt removed from the cavity, were poured into the cavity. The temperature of the liquor was raised

Table 3. Chemical Composition of Gas Produced During Run 15

8 M Purex, Acid, No Salt Added

Radiation field, 3.5×10^6 r/hr

Irradiation Time (hr)	Sample Volume (cc)	Composition (%)				
		Cl_2	NO_2	O_2	H_2	N_2
0.5	59.5	1.2	1.6	21.6	0.3	75.2
2.25	98.7	0.9	0.2	20.6	0.0	78.3
4.75	97.0	0.4	0.2	20.9	0.1	78.4
17.25	96.2	0.4	0.2	21.8	2.9	74.6
22.25	67.8	0.4	0.9	24.9	1.2	72.6
28.25	81.9	1.8	1.7	26.3	2.3	67.9
41.25	56.5	1.2	1.4	25.0	2.3	70.2
48.60	43.3	1.6	1.0	25.5	3.0	69.1

Table 4. Chemical Composition of Gas Produced During Run 10B

8 M Purex, adjusted to pH 10, saturated with salt

Radiation field, 3.5×10^6 r/hr

Irradiation Time (hr)	Sample Volume (cc)	Composition (%)				
		Cl_2	NO_2	O_2	H_2	N_2
0.83	60.3	0	0	19.6	0	80.4
3.83	97.0	0	0	21.1	0	78.9
5.58	99.0	0	0	28.2	0	71.8
8.33	95.8	0.5	0	28.9	0.6	69.9
11.33	94.3	0	0	34.0	0.5	65.4
16.33	94.8	0	0	41.0	1.2	57.9
20.33	80.3	Trace	0.5	41.5	1.6	56.3
26.33	39.8	0	0.2	53.0	2.3	44.5
29.33	68.4	0.3	3.8	57.2	3.6	35.1
35.33	98.2	Trace	1.1	70.1	3.6	25.0
43.33	94.8	0	7.5	59.4	3.9	29.3
46.58	58.9	0	13.7	41.5	1.4	41.5
53.33	88.7	0	3.2	70.1	4.5	22.2
66.33	97.6	0	5.0	70.7	4.0	20.3
70.75	96.2	0	1.0	44.1	1.9	52.6

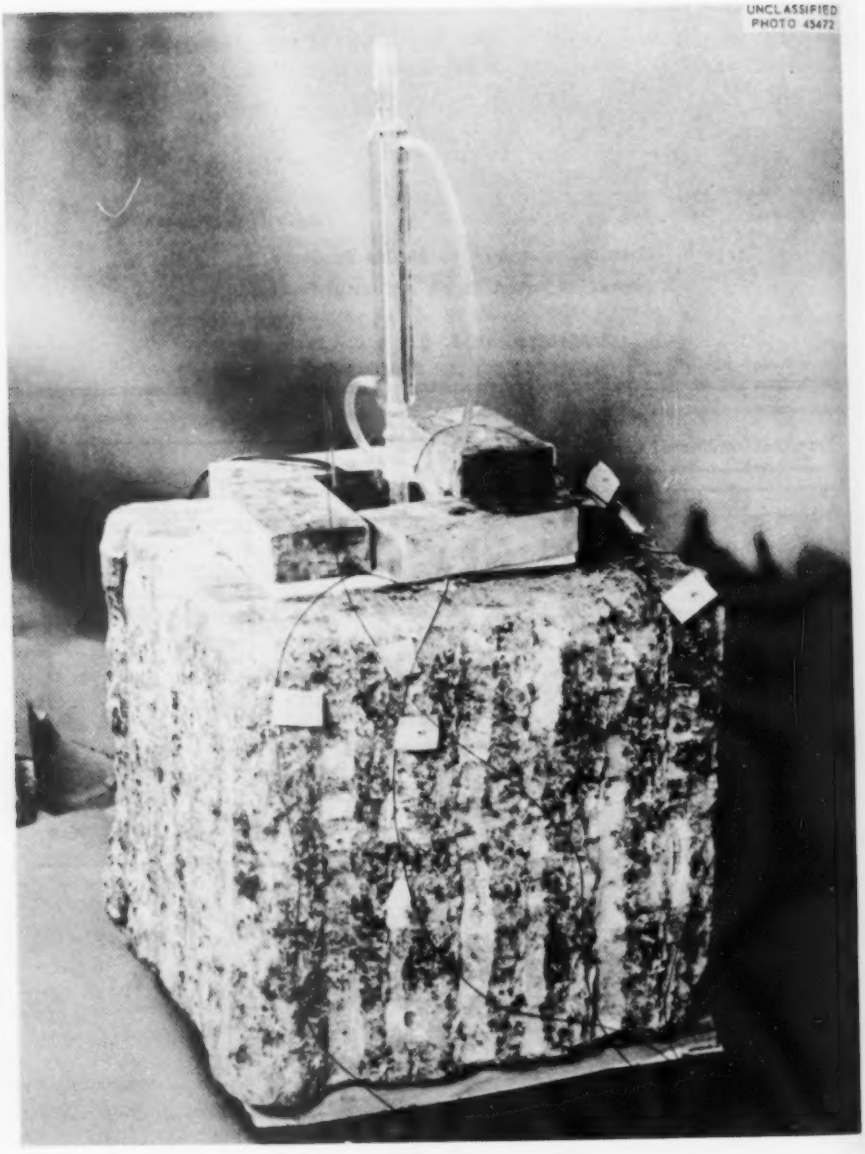

Fig. 15. Model of Field Experiment.

Table 5. Thermocouple Locations

TC	X*	Y	Z
1	16	3	7 1/2
2	16	10	10
3	10	5	8
4	10	3	11
5	4	5	11 1/2
6	4	5	8
7	10	12	11
Block	0 - 20	0 - 20	0 - 20
Cavity	6.25 - 13.75	0 - 9.5	6.25 - 13.75
Heater	10	4 - 7	10

* X, Y, and Z equal zero at top left front corner of block.

to 90° C. The recorded temperatures of thermocouples 3 and 4 (Fig. 16) show a small thermal gradient in the liquor. Evidently the convective currents are not sufficient to equalize the temperatures throughout the liquor. The maximum temperature recorded out in the block was 50° C. Temperatures in various parts of the block show that there is no difference in the thermal conductivity-heat flux ratio in any part of the block.

During the course of the experiment recrystallized salt appeared in No. 2 and No. 5 thermocouple holes at the outside surface of the block. The recrystallized salt also appeared at various other locations on the surface. Salt continued to crystallize, and the growths increased during the course of the experiment. The growths occurred both above and below the liquid level in the cavity. After two months the block was permitted to cool, the cavity equipment and wastes were removed, and then the block was broken apart for examination. Five liters of the original six liters of waste solution were recovered. The upper 2 1/2 inches of the cavity was enlarged significantly from its original dimensions (Fig. 17).

A thin layer of hydrous iron oxide (< 1/16 inch thick) from the neutralized wastes coated the lower 7 inches of the cavity walls below the liquid level. Apparently, the wastes did not penetrate this coating. A layer of recrystallized salt about 2 inches thick "bridged" the cavity above the liquid level. Salt, in the upper 2 1/2 inches of the cavity immediately below the cover, was dissolved by the condensing water vapor and was recrystallized as the brine moved down the exposed side walls of the cavity to form a firm bed of crystals over the liquor.

TEMPERATURE (°C)

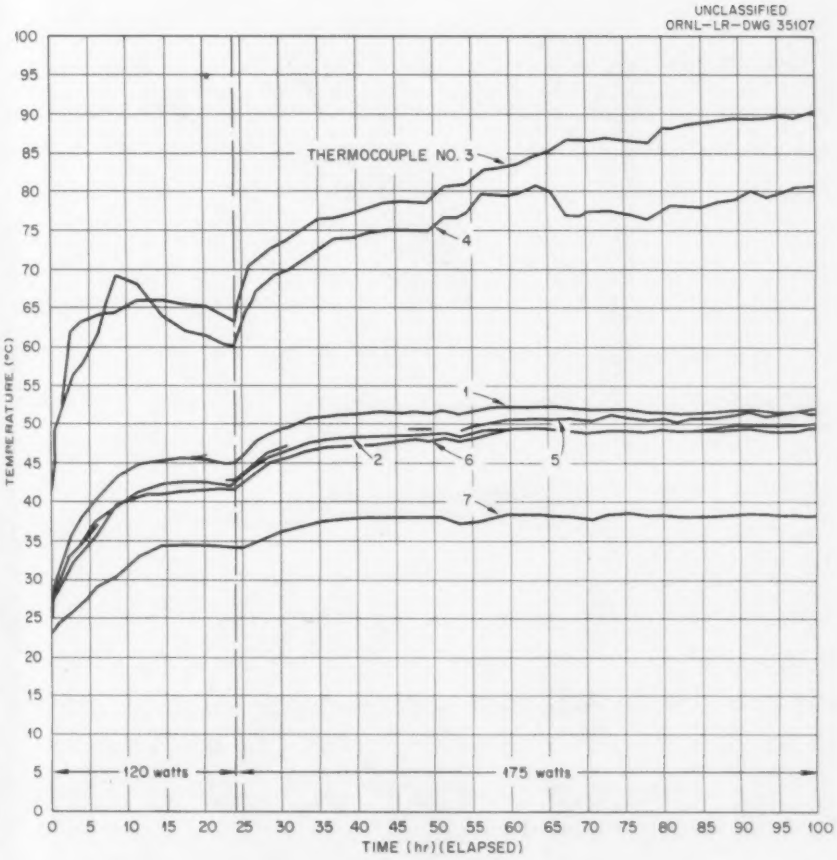

Fig. 16. Temperature Distribution in Salt Block.

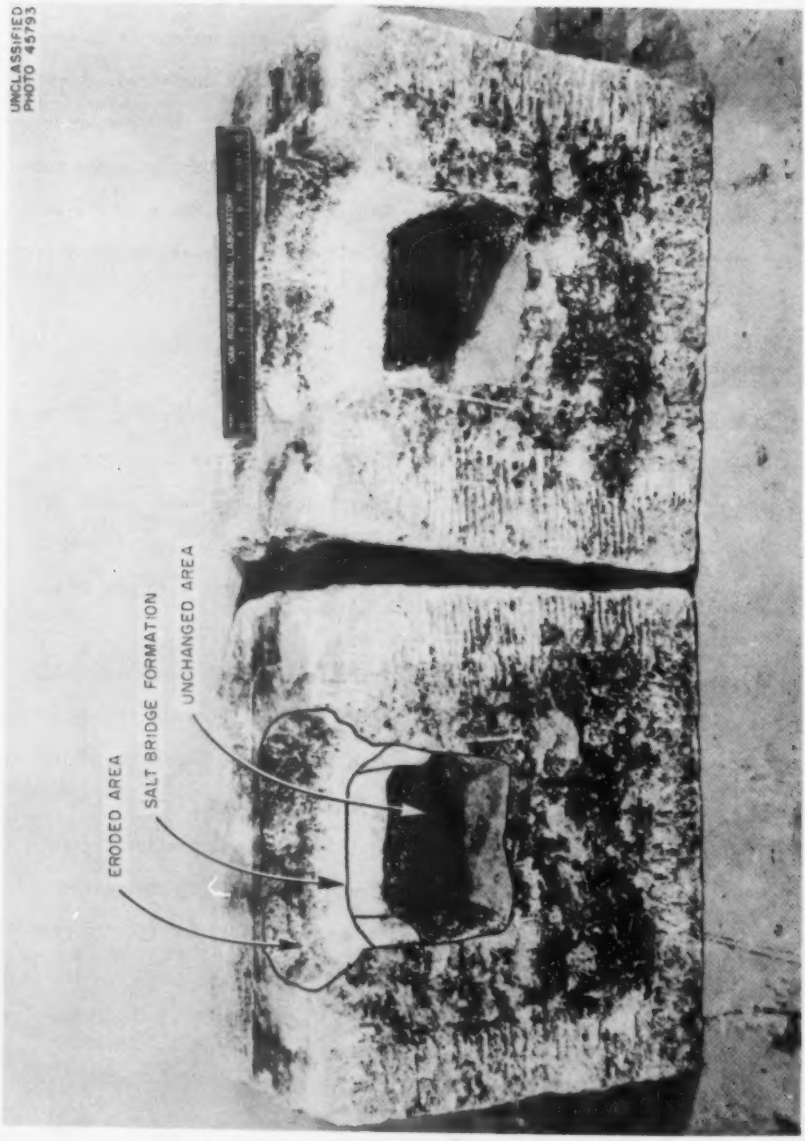

Fig. 17. Results of Tests on Model of Field Experiment.

FIELD TESTS

In the laboratory, it is practically impossible to reproduce the conditions that exist in a natural rock salt formation. Samples removed from the formation go through a process of stress relief, violent enough in some cases to shatter the sample. Even if the samples are not shattered, there is some alteration of the physical properties of the rock. If the overburden pressure is reapplied, the stresses induced will not necessarily be the same unless the sample is completely elastic, which is not true for geologic materials. Therefore, it is necessary to conduct field experiments before a prototype disposal facility can be designed.

Experimental Design

Information can be obtained in the field experiment regarding (1) the structural integrity of the cavity; (2) the migration of nuclides from the cavity; (3) the migration of the cavity itself, due to the condensation of the water vapor on the surfaces causing the salt to go into solution and to flow back into the waste cavity; (4) temperature gradients, both inside and outside the cavity; and (5) gas production.

Eight tests are planned requiring eleven excavations. Each test will run for approximately six months. Since some tests will run concurrently, the over-all field test program will require about two years. Major variables to be studied include: (1) the type of waste (Purex and acid aluminum nitrate), (2) the concentration and pH of the wastes, (3) the geometrical configuration of the cavity, (4) the surface area/volume relationships, (5) the method of construction (hand excavated, blasted, or solution-mined), and (6) the rate of

Fig. 17. Results of Tests on Model of Field Experiment.

heat production.

Site Selection

The first tests with synthetic wastes and simulated fission product decay heating will be followed by tests with actual radioactive wastes. These preliminary nonradioactive tests will be conducted in mined-out space in some salt mine.

During the past year, a Laboratory subcontractor (Geotechnical Corporation, Dallas) investigated operating salt mines in the northeastern section of the country. The investigation was limited to this area, because bedded deposits are found in this area, and it seemed likely to be the general location of the first commercial fuel-reprocessing plant. It was found that the owners were amenable to the use of their mines for the field tests, since the first tests would not involve the use of radioactive wastes. There would be a number of advantages in using mined-out space for these tests, e.g., the immediate availability of a test site, a competent work force with the necessary equipment available to excavate the cavities, and the necessary auxiliaries - shaft, hoist, electrical power, railroad sidings, etc.

The Carey Salt Company (Hutchinson, Kansas) and International Salt Companies (Detroit, Michigan) expressed their willingness to participate in these tests. Their mines satisfied most of the requirements of a good test site. A detailed analysis of the geology, geographic location, facilities in the mine, estimated costs, and the possible location of future tests indicated that the Hutchinson mine would be more desirable.

Accordingly, the Laboratory entered into contractural arrangements with the Carey Salt Company for the use of an unoccupied section of their mine to

carry out these experiments. The first tests will begin in July, 1959.

Engineering Design

The proposed test facility is shown in Fig. 18. The cavity is 7 1/2 ft square and 10 ft deep, a size that will give a surface area equal to the surface area of a 10-foot diameter sphere, when the cavity is filled to the 7 1/2-foot level. Thus, if thermal data from the field experiment agree with theoretical heat calculations, the scaling factors can be computed. Jakob has shown that the thermal distribution for small spheres and small cubes is the same.[12] The facility includes the cavity, immersion heaters, a gas tight cover, and measuring devices and recorders. All data, with the exception of liquid level, will be on strip-chart recorders. An alarm system set to function on high temperature, moisture penetration, high pressure, and loss of electrode power will be installed in the experimental area and telemetered to the mine office.

All equipment will be prefabricated to sizes that can be brought down the shaft and assembled in the mine. The cavity will be excavated in the floor of an existing room in the mine. Conventional room and pillar mining has been used in the Hutchinson mine, the pillars being 50 ft by 50 ft and the rooms, 50 ft by 50 ft. The floor-to-ceiling height varies from 10 to 12 ft. In older sections of the mine, the rooms are 50 ft by 300 ft and with 6-foot-high pillars, 20 ft by 300 ft. Core drilling in the proposed test area of the mine has shown that the salt bed beneath the present floor exceeds 30 ft in thickness.

The first two cavities will be hand-excavated to avoid the shattered zones caused by blasting. Blasting operations appear to disturb and shatter the rock to a depth of 12 to 18 inches.

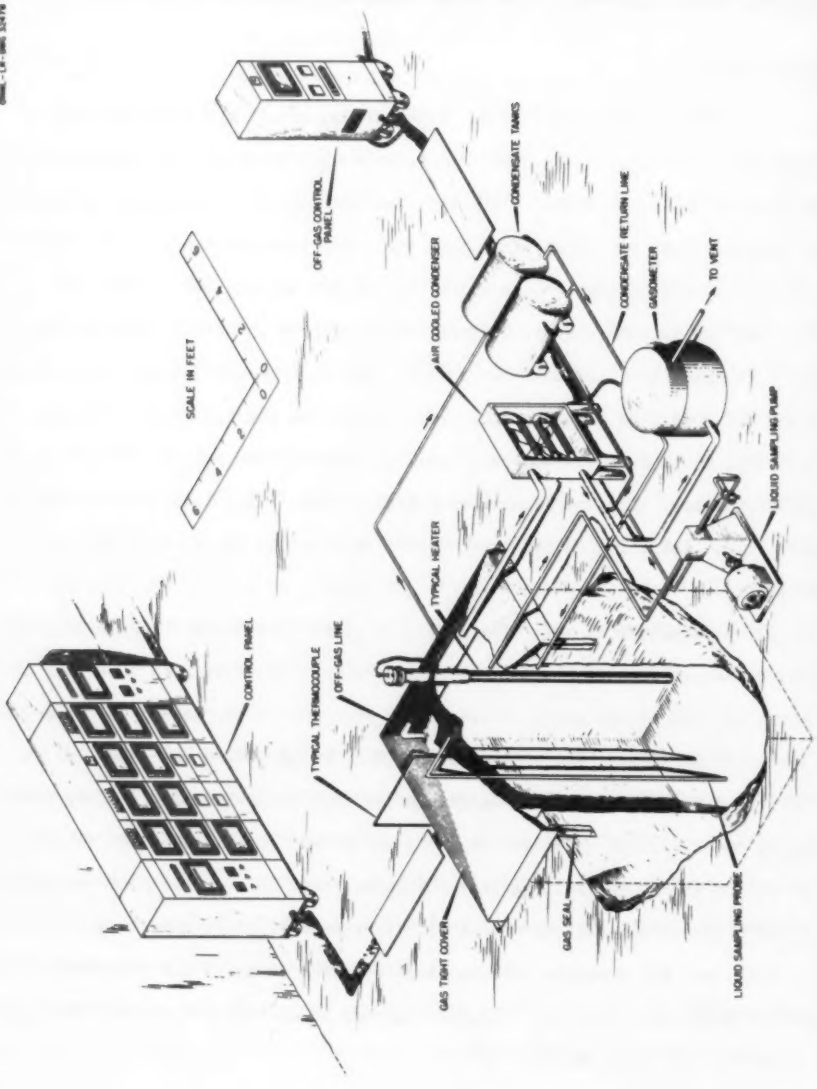

Fig. 18. Proposed Salt Field Experiment.

Conductivity probes will be placed next to the cavity to measure any moisture changes in the salt. The electrical conductivity of salt is extremely sensitive to changes in moisture content; so the changes in electrical conductivity will indicate any movement of the wastes out into the surrounding salt. A gas tight cover will be installed so that off-gases can be collected, condensed, measured, analyzed, and returned to the cavity. As discussed previously, the total quantity of gas produced will be small.

Heat must be provided to simulate the heat of radioactive decay. It is, of course, impossible to duplicate exactly the heat generated by fission-product decay where each nuclide produces heat as a function of its energy and half life. However, the calculated temperature rise is based on the assumption that the waste is uniformly mixed, so uniform heat production is desired. In the case of neutralized wastes, more fission products are in the precipitate than in solution, so additional heat must be distributed in the precipitate. Thermocouples will be placed in the cavity (in both liquid and gaseous phases) and in the salt surrounding the cavity.

Fig. 18. Proposed Salt Field Experiment.

Operating Procedure

After completion of construction and pretesting, about 7,000 gal of synthetic waste (saturated with salt) will be transferred to the two test cavities. When the waste solution temperature has reached the ambient mine temperature, the heaters will be turned "ON," and the power varied to correspond to a predetermined fission product decay function (representative of a given waste type) while considering the decreasing resistivity of the solution with increasing temperature.

Temperatures in the cavity (above and below the liquid level) and in the salt surrounding the cavity will be recorded continuously. Also, heater power and electrical conductivity of the surrounding salt will be recorded continuously. Periodic samples of cavity solution, condensate and off-gases will be withdrawn and analyzed. Charts from the recorders will be removed and replaced as needed and routine preventative maintenance of the recorders and power controls will be performed at regular intervals.

The peak temperature of the cavity solution ($\sim 80^{\circ}$ C) is expected to occur in approximately five months. Once the solution temperature levels off, the heaters will be turned "OFF," and the solution will be cooled to ambient temperature. After removal of the cavity cover and heaters, the wastes will be disposed of in a nearby solution cavity. The empty cavities will then be filled with rock salt and the equipment moved to the next test cavities.

MATERIALS STUDY

A critical design problem has been that of finding suitable materials for the fabrication of heaters, cavity covers, and off-gas equipment because of the corrosiveness of waste-salt mixtures and the associated off-gases. In the actual storage of radioactive wastes, this will not be a serious problem.

Corrosion Studies

The wastes themselves are not highly corrosive. Both acid aluminum nitrate and Purex (nitric acid) wastes are readily handled in austenitic stainless steels (300 series) or, when neutralized with sodium hydroxide, in ordinary steel. However, when the acid solutions are saturated with common salt (NaCl),

the resulting solutions and vapors are extremely corrosive. The high corrosion rates in the nitric acid-sodium chloride solution are due to the presence of the same ions as are present in a nitric acid-hydrochloric acid (aqua regia) solution. Due to the presence of nascent chlorine, aqua regia is one of the most corrosive materials known, readily dissolving both platinum and gold, as well as normal metallic materials. Corrosion rates significantly greater than 0.001 inches/year are considered to be unsatisfactory.

Table 6 presents the results of a study of possible construction materials. The waste of immediate interest is Purex waste under acid conditions, since this is the most abundant present-day waste. The table shows that only a limited number of materials are suitable. Titanium is considered the most resistant material on the basis of corrosion tests carried out at Battelle Memorial Institute in connection with Darex process development.[13] These were tests in HNO_3 acid containing HCl, or with HCl gas bubbling through the liquid. A second metal which shows promise is a cobalt base alloy, Haynes 25 (Haynes Stellite Co.). The corrosion rate was negligible in 77 days at 80° C with the sample suspended at the interface of the waste and vapor under reflux conditions. There was no evidence of pitting. Other samples containing welds showed higher corrosion rates.

The nonmetallic, inorganic materials considered include graphite and glass-lined steel. Graphite is brittle and involves some limitations in fabrication, since it cannot be welded. A glass lining applied to a cover fabricated of steel would be satisfactory under acid conditions. However, glass and other siliceous materials are appreciably attacked by strong NaOH present in the neutralized waste.

Mong organic materials, Teflon (E. I. Du Pont de Nemours and Co., Inc.) appears to be completely resistant. The cost of the material, combined with

Table 6. Corrosion Data in Salt Saturated Simulated Waste

Material	Corrosive Medium (salt saturated)	Temperature °C	Exposure Days	Corrosion Rate (in./yr)	Remarks
410 SSt	Hope	Approx. 22°	238	----	Failed.
410 SSt	Neut. Hope	Approx. 22°	238	Negl.	
410 SSt	Purex-7MHNO$_3$	Approx. 22°	238	----	Failed.
410 SSt	Purex-Neut.	Approx. 22°	238	Negl.	
316 SSt	Hope	Approx. 22°	238	Negl.	
316 SSt	Hope-Neut.	Approx. 22°	238	Negl.	
316 SSt	Purex-7MHNO$_3$	Approx. 22°	238	----	Failed.
316 SSt	Purex-Neut.	Approx. 22°	238	Negl.	
347 SSt	Hope	Approx. 22°	238	0.0025	
347 SSt	Hope-Neut.	Approx. 22°	238	Negl.	
347 SSt	Purex-7MHNO$_3$	Approx. 22°	238	----	Failed.
347 SSt	Purex-Neut.	Approx. 22°	238	Negl.	
Inconel	Hope	Approx. 22°	238	0.013	
Inconel	Hope-Neut.	Approx. 22°	238	Negl.	
Inconel	Purex-7MHNO$_3$	Approx. 22°	238	----	Failed.
Inconel	Purex-Neut.	Approx. 22°	238	Negl.	
Inor	Hope	Approx. 22°	238	0.008	
Inor	Hope-Neut.	Approx. 22°	238	Negl.	
Inor	Purex-7MHNO$_3$	Approx. 22°	238	----	Failed.
Inor	Purex-Neut.	Approx. 22°	238	Negl.	
Hastelloy C	Purex-7MHNO$_3$	Approx. 22°	139	0.0004	
Hastelloy C	Purex-7MHNO$_3$	Approx. 80°	19	----	Failed.
Haynes 25	Purex-7MHNO$_3$	Approx. 80°	77	0.0007	Stamped sample.
Haynes 25	Purex-7MHNO$_3$	Approx. 80°	34	0.00147	Sample cut from sch. 40 pipe.
Haynes 25	Purex-7MHNO$_3$	Approx. 80°	34	0.004	Sample from sch. 40 pipe containing butt weld.
Haynes 25	Purex-Neut.	Approx. 80°	77	Negl.	Stamped sample.
Titanium	Purex-7MHNO$_3$	Approx. 80°	63	0.0008	
Titanium	Purex-Neut.	Approx. 80°	63	Negl.	
Titanium	Purex-7MHNO$_3$	Approx. 22°	127	Negl.	
Teflon	Purex-7MHNO$_3$	Approx. 100°	70	----	No change. Slight weight gain.

Table 6. Corrosion Data in Salt Saturated Simulated Waste (continued)

Material	Corrosive Medium (salt saturated)	Temperature °C	Exposure Days	Corrosion Rate (in./yr)	Remarks
Polyvinyl Chloride (unplasticized)	Vapor above Purex-7MHNO$_3$	Approx. 80°	24	Negl.	Approx. 1.4% weight gain without apparent swelling.
Polyvinyl Chloride	Purex-7MHNO$_3$	Approx. 80°	21	Negl.	Totally immersed in medium. One per cent gain without apparent swelling.
Graphite	Purex-Neut.	Approx. 80°	30	----	No corrosion. Slight weight gain.
Graphite	Purex-7MHNO$_3$	Approx. 80°	30	----	No change. No weight loss during use of graphite as electrode in heating experiments.

1. All samples exposed partially immersed, except as noted.
2. Samples at 80° C (and higher) under refluxing conditions.

the problems of fabricating a large cover assembly (approximately 10 ft by 10 ft by 3 ft high) makes it unattractive. The possibility of bonding thin Teflon to a steel cover is being studied. Unplasticized polyvinyl chloride may be suitable for some of the experimental equipment though it exhibits some swelling in the acid liquid.

Heater System

Uniform heat production in the solution is desirable in the field cavities. It may be achieved by covering opposite walls of the cavity with flat plate electrodes and applying a voltage between them. The resulting current flow through the solution (electrolytic heating) will thus produce uniform heating. If the two plates are perforated and moved away from the walls slightly, the solution will have free access to the walls, producing a uniform temperature throughout.

Tests indicate that conventional immersion heaters in the acid waste would not withstand the corrosive conditions. Corrosion is not a serious problem in neutralized waste, but because of viscosity of the neutralized precipitate, thermal convection currents cannot be relied upon to equalize the heat distribution. Laboratory experiments indicate that a reasonably uniform heat distribution in the precipitate cannot be obtained readily with immersion heaters. Therefore, the wastes will be heated electrolytically.

Electrolytic heating introduces problems of (1) the design of the electrodes, (2) the design of the power supply, and (3) possible gassing due to electrolysis.

Electrodes.-- Almost any corrosion resistant material should be suitable for use as electrodes in the neutralized waste solution, since neutralized

waste is not very corrosive. However, the choice of materials for use in the acid waste is limited. Carbon, or graphite, seem to be the most desirable from the standpoint of corrosion resistance, as well as cost and availability. However, graphite is rather brittle and would be fragile in a perforated plate of the proper dimensions.

Since the salt-saturated acid waste has a fairly low viscosity it would be expected that thermal convection currents would tend to produce a uniform temperature throughout the solution, even with nonuniform heating. Several arrangements of cylindrical graphite electrodes were tried on a scale of one inch to the foot in an attempt to produce uniform heating. The electrode configuration adopted produced the power (rate of heat production) distribution shown in Fig. 19. When the salt solution temperature was raised 10° C by this electrode arrangement the temperature variation to within one-half inch of the walls and the electrodes was only one degree, indicating a relatively uniform heat distribution.

Power Supply.-- The electrical resistivities (reciprocal of conductivity) of the salt-saturated acid waste, the neutralized precipitate, and the neutralized supernate measured by an Industrial Instruments Company Conductivity Bridge are shown in Table 7. It may be observed that the neutralized waste has a resistivity at 20° C, nearly three times that of the acid waste, but at 80° C the ratio drops to only a little greater than two. Thus, the voltage and current range of the power supply required to heat the neutralized waste will be greater than that required by the acid waste.

The electrical resistivities of both the acid and the neutralized wastes are relatively low and, to produce the desired power in the solution, will

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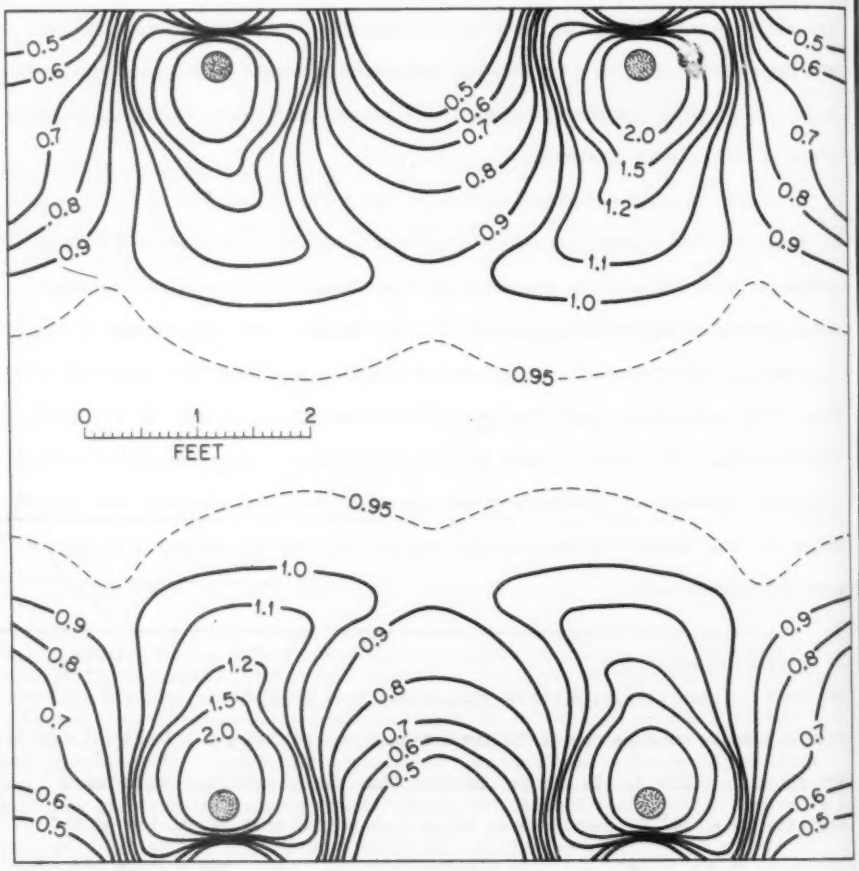

CONSTANT POWER PRODUCTION CONTOURS
ARBITRARY POWER SCALE

Fig. 19. Power Distribution with Four Electrodes.

Table 7. Waste Solution Resistivity

Solution	Resistivity (ohm-cm)	
	20° C	80° C
Salt Saturated Acid	2.4	1.2
Salt Saturated Neutralized Precipitate	6.8	2.6
Salt Saturated Neutralized Supernate	6.2	2.4

require a high-current, low-voltage power supply. Since the resistivities of the solution drop rapidly with increasing temperature, decreasing voltage and increasing current will be necessary in order to maintain a given power input. Fission product power production decreases with time and will tend to reduce the increased current demand caused by increasing temperature, but will call for a further decrease in voltage.

The initial voltages required for the two types of waste, however, are not directly indicated by the ratio of their 20° C resistivities, due to two factors. One is that the use of carbon rod electrodes in the cavity filled with acid waste will produce a total effective solution resistance which is greater than that which would be obtained with plate electrodes, thus requiring a greater voltage to produce the same power. The other factor is that from 70 to 90% of the fission product radioactivity in the neutralized waste will be in the precipitate, thus requiring most of the power in the precipitate. To meet this requirement, the plate electrodes will be in the precipitate only and not extend up into the supernate. This results in a greater effective resistance and consequent increase in required electrode voltage.

Tests using carbon electrodes in approximately two and one-half gallons of salt-saturated acid waste at several temperatures and power inputs showed that the wattage input was essentially the same as the volt-ampere product, and thus there is no reactive current capacity which must be built into the supply.

The Hutchinson mine has 2300 volt, 60 cycle, 3 phase power available. This will be stepped down in voltage, and each experiment will be connected to a separate phase, since the electrodes need to run on single phase power to prevent sacrifice of the graphite.[14] One way to obtain the necessary flexibility is to step down to 440 volts before feeding into a Powerstat or other

variable transformer and then, with a special transformer, step down further to the voltage necessary at the electrodes.

The approximate voltage and current ranges for the acid waste experiment will be from 25 volts and 800 amperes at 20° C to 10 volts and 2000 amperes at 80° C. For the neutralized waste the approximate range will be 40 volts and 500 amperes to 15 volts and 1350 amperes.

Further experiments are planned in order to obtain accurate figures for scale up calculations on the power supply before equipment is obtained.

Gassing.-- The passage of an alternating current through a conducting solution will produce gas at the electrodes if the current density at the electrodes is great enough. Tests of carbon electrodes at current densities greater than those that will be encountered in the field experiment have shown no visible gassing.

Further tests will be run to establish the actual gas production rate, if any, so that a correction may be made in the off-gas analysis of the field experiments.

CONCLUSION

This status report has described analytical work on the thermal problem associated with disposal of radioactive wastes in salt, the experimental studies of the effect of radiation on the structural properties of rock salt, and the chemical interactions between wastes and salt. The selection of a test site, the design of the field test, and the material studies associated with the field tests are also described.

The results of the work have been applied to the design of a series of tests leading to the final test, actual disposal of radioactive waste in a salt cavity. Theoretical calculations and laboratory scale experiments with disturbed samples can only give us an approximation of the results to be expected in the field. Similarly, the preliminary field tests can only simulate heat generation and chemical compatibility. The definitive test will be the actual disposal of radioactive wastes under carefully controlled conditions in some remote and isolated salt cavity.

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Thermal Considerations *in* Deep Disposal of Radioactive Waste

by

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SUMMARY

The following pages contain some calculations and arguments relative to the thermal problems in disposing of large quantities of radioactive waste in geologic formations. The Committee on Waste Disposal has recommended that research be undertaken to determine feasibility of disposing of wastes in salt beds and salt domes, and in the porous strata of deep synclinal structures with complete closure. The experimental approach is discouraging because of the long time required for the heating to reach its maximum, of the order of 40 years or more, with the possibility that important consequences may not be observable for longer times. The uncertainties arising from uncontrollable underground conditions must be balanced against the apparent economies of the proposed disposal methods; alternatives such as storage in surface tanks or in solidified form, though possibly more costly, may offer greater opportunity for monitoring and control.

The only aspect of the problem of safety which is considered in this report has to do with the generation of heat in the waste; this is treated in terms of simplified, undoubtedly artificial models, which may nevertheless retain the principal features of the real situations, so long as the temperatures remain within some poorly-understood limits. The limiting conditions are then examined. The results suggest that nearly any quantity of waste producing heat at the rate of 0.01 watts per gallon, if distributed in a layer of the order of 100 meters thick, can be accommodated without undue rise of temperatures. Concentrations much above this level would raise questions exceedingly difficult to answer.

It does not appear that any decisive advantage is gained by burial at excessive depths (greater than a few thousand feet, for example) provided that potable water supplies and other natural resources are adequately protected. Some real geologic formations which are potential reservoirs should be studied in detail with respect to lithology, stratigraphy, structure, proximity to cities or towns, nature of contained water, relations to potable water, etc. A major difficulty comes in deciding when and how the reservoir condition will break down as the content of the reservoir heats up.

DISPOSAL IN POROUS STRATA

We wish to specify an upper limit for the concentration of liquid wastes which, under given conditions, would not lead to undesirable thermal effects.

There are two problems here, which can to some extent be treated separately. First, we have to make estimates, as realistic as possible, of the rise of temperature, as a function of configuration, concentration and time. Second, we have to decide at what point the assumptions made for the first part will break down—in other words, to decide when new effects will begin. This second part is the more difficult: It is relatively easy to compute the rise of temperature so long as the principal agency of heat loss remains conduction; it is not so easy to decide when convection may begin to mix waste with the surrounding ground water, or what the consequences may be.

Once material has been injected into a porous formation, it is subjected to a variety of disturbing processes. Chemical effects may fix some of the elements near the point of entry; fractures and sedimentary features of primary origin may permit migration in unexpected directions, etc. Experiments bearing on the possibility that the radioactive ions would be fixed in disproportionate amount close to the point of entry are evidently important, though it is likely that local hot spots would be cooled by the development of convective movements of the fluid.

Temperatures in a Heated Layer

For the purpose of making preliminary estimates, it will be supposed below that it is possible to distribute the material uniformly in a horizontal layer of uniformly porous rock, in a layer which is thin by comparison with its horizontal dimensions, and that the material is confined within this layer by impermeable boundaries. It is further supposed, as most closely resembling the probable real circumstances, that circulation within this layer produces a virtually uniform temperature across the layer, and that loss from the layer is entirely by conduction. A relatively thin layer of wide extent is probably the most favorable configuration for combining high heat loss with large volume. Table 1 shows the capacity, per km^2, of beds of various thickness for an average porosity of 20 percent.

TABLE 1

CAPACITY PER SQUARE KILOMETER
OF LAYER OF 20 PERCENT POROSITY

Thickness meters	Liters millions	Gallons millions
1	200	53
10	2,000	530
100	20,000	5,530

The heat capacity of the medium will be intermediate between that of water, 1 cal/cm^3 deg, and that of ordinary rock, about 0.5 cal/cm^3 deg. For rock of pore volume x (fractional) filled with water, the mean heat capacity is then 0.5 (1 + x) cal/cm^3 deg; this varies from 0.5 for x = 0 to 0.7 for x = 0.4 (40 percent).

The radioactive heat sources after a reasonable cooling period being principally Sr^{90} and Cs^{137}, we take as an approximate single decay constant a half-life of about 30 years, or a disintegration constant of $\alpha = 0.02$ per year. If heating is according to $A_o e^{-\alpha t}$, in cal/cm^3 year, then the total heat generated for all time is A_o/α or $50A_o$, and of this, half is produced in the first 30+ years. Suppose that the rate of heating of the waste is W, in watts/gal. This is equivalent to 2000 W, in cal/cm^3 year. With a porosity of 20 percent, we have $A_o = 400W$ cal/cm^3 year, and the ultimate rise of temperature, if no heat were lost, would be 33,000 W degrees. As the following calculations will show, a considerable fraction of the heat will be lost, but it is immediately suggested that W must not exceed values of the order of 0.01 (1 watt per 100 gallons).

For W = 0.01, A_o = 4 cal/cm^3 year (porosity 20 percent). A layer 10 cm thick thus produces 40 cal/cm^2 year, which is about the normal geothermal flux. A layer 1 meter thick with these values produces 10 times the normal flux. These figures need not cause alarm without further examination, but clearly we are dealing with important sources of heat. The heat flow from the intensely active Wairakei area of New Zealand is believed to amount to about 150,000,000 cal/sec; the area involved is somewhat uncertain, but if we take 20 km^2, the "normal" heat flow would be about 250,000 cal/sec. The actual heat loss is then about 600 times normal. This intensity would be produced, at least for a period of the order of 30 years or so, by a layer 6000 cm or 60 meters thick, porosity 20 percent, filled with waste having a concentration of 0.01 watt/gal.

In the following, we shall neglect the effect of the Earth's surface on the cooling of the layer. This is permissible as soon as the depth to the layer exceeds a few hundred meters, for times of the order of a few hundred years.

Only a single injection of waste is considered; this is supposed to form a layer and to remain in place, except for stirring within the layer. With a little more trouble, calculations for several discrete injections, or for a continuous introduction of material could of course be made.

The problem in heat conduction to be treated is as follows: in the infinite medium, the layer, $-a<x<a$, is heated uniformly at the rate $A_o e^{-\alpha t}$, in cal/cm^3 year. Within the layer, the temperature is uniform. The initial temperature of medium and layer is zero. The conductivity of the medium is K, heat capacity per unit volume, c; the heat capacity of the layer, per unit volume, is c' let $n = c/c'$. Because of the symmetry about the plane x = 0, we need consider only positive values of x. Then first considering constant heating, A, the equations are:

$$\text{for } 0<x<a, \quad ac' \frac{\partial T}{\partial t} = Aa + K \frac{\partial T}{\partial x}\Big|_{x=a} ,$$

$$\text{for } x>a, \quad c \frac{\partial T}{\partial t} = K \frac{\partial^2 T}{\partial x^2} .$$

Applying the Laplace transformation, these become

$$\bar{v} = A/c'p^2 + (K/ac'p) \frac{\partial \bar{v}}{\partial x}\Big|_a \quad \text{and} \quad \frac{\partial^2 \bar{v}}{\partial x^2} = q^2 \bar{v} ,$$

where $\bar{v}$ is the transform of T, $q^2 = p/k$, $k = K/c$. The desired solution for $x>a$ is $\bar{v} = Be^{-qx}$. Then $\frac{\partial \bar{v}}{\partial x}\Big|_a = -qBe^{-qa}$. The temperature must be continuous at $x = a$; this determines B:

$$B = \frac{Ae^{qa}}{c'p^2} \frac{qa}{qa + n} = \frac{Ae^{qa}}{kc'} \frac{1}{pq(q + h)} , \text{ with } h = n/a .$$

From this we have,

$$\text{for } 0<x<a, \quad \bar{v} = \frac{An}{K} \frac{1}{pq(q + h)}$$

$$\text{for } x>a, \quad \bar{v} = \frac{An}{K} \frac{e^{-q(x - a)}}{pq(q + h)} .$$

This is now in the form listed in the table of transforms in Carslaw and Jaeger (1947, p.381, 15) and the temperature may be written at once as

$$T = \frac{Aa^2}{Kn} \left\{ \sqrt{\frac{4kn^2t}{\pi a^2}} - 1 + e^{kn^2t/a^2} \operatorname{erfc} \sqrt{kn^2t/a^2} \right\}, \quad \text{for } 0<x<a .$$

The result for exponential decay of the heating rate is readily obtained from this by applying Duhamel's theorem, and we find

$$T = \frac{A_o}{c'\alpha} \frac{e^{-\alpha t}}{(1 + kn^2/a^2\alpha)} \left\{ e^{\alpha t(1 + kn^2/a^2\alpha)} \operatorname{erfc} \sqrt{kn^2t/a^2} - 1 \right.$$
$$\left. + \frac{2}{\sqrt{\pi}} \sqrt{\frac{kn^2}{a^2\alpha}} \int_0^{\sqrt{\alpha t}} e^{z^2} \, dz \right\}, \quad \text{for } 0<x<a .$$

All of the functions in this expression have been tabulated and numerical results may be calculated for any set of assumptions.

Two examples have been worked out, with the following choices of constants: Ex. 1, a = 40 meters = 4000 cm; k = 0.01 cm²/sec = 32.10^4 cm²/year; α = 0.02 per year. We then have $k/\alpha = 16.10^6$ cm², and $k/\alpha a^2 = 1$. For simplicity, n is taken as 1; this will be reasonably correct if the rock surrounding the layer is also saturated and has about the same porosity. Ex. 2, a = 10 meters = 1000 cm; other values the same as in example 1.

There are various ways of exhibiting the results; perhaps the most flexible is to show the coefficient of A_o/c' as function of the time after injection. This coefficient is also a time, and will be given in years. The rise of temperature is given by A_o/c', which is the rate of rise at zero time, multiplied by this coefficient, which gives, as function of time, an equivalent heating period at the initial rate. For comparison, the function $(1 - e^{-\alpha t})/\alpha$ is also shown; this, multiplied by A_o/c', gives the rise of temperature if no heat is lost. It approaches $1/\alpha$ as an asymptote, in this case, 50 years.

We let the rise of temperature be given by $A_o f(t)/c'$. The function f(t) is given in Table 2 for the two examples, and for the case of no heat loss, which is the limiting case for a very thick layer. It is also instructive to form the ratio R(t) = f(t) divided by $(1 - e^{-\alpha t})/\alpha$; this gives the fraction of the heat generated up to time t which is represented in the rise of temperature at that time. The function 1 - R(t) then gives the fraction of the generated heat which has been lost by conduction.

For the 20-meter layer, a flat maximum of temperature is reached after about 50 years; for the 80-meter layer, the maximum is reached later, at about

TABLE 2

t years	f(t), years			R(t)	
	20 m	80 m	inf.	20 m	80 m
2	1.20	1.70	1.96	0.612	0.867
12.5	4.10	7.90	11.06	.371	.714
32	6.01	14.06	23.64	.254	.595
50	6.47	16.68	31.60	.205	.528
84.5	6.19	17.96	40.78	.152	.440
112.5	5.65	17.48	44.73	.126	.391
144.5	5.01	16.41	47.23	.106	.347
200	4.12	14.38	49.08	.084	.293

80 years, and is 2.77 times as high, concentrations being equal. Since, under these conditions, there is 4 times as much waste in the 80-meter layer, the thicker layer is more efficient in restricting the rise of temperature. If the concentration in the 20-meter layer were 4 times as great as in the 80-meter layer, so that the content per unit area was the same, then the maximum temperature in the 20-meter layer would be about 45 percent higher than the maximum of the 80-meter layer. This is a consequence of the greater thermal mass of the thicker layer, which retards the rise of temperature while radioactive decay is reducing the rate of heating. The maximum is reached later, and is relatively lower than for the same amount in a thinner layer.

With the figure adopted earlier (porosity 20 percent $\alpha = 0.02$ per year) we had $A_o = 400$ W cal/cm^3. year, W = watts per gallon. $A_o/c = 670$ W degree/year ($c = 0.6$ cal/cm.deg.). This led to ultimate temperatures of 50 x 670 = 33,000 W if no heat were lost. The maximum for the 20-meter layer corresponds to an effective time of 6.5 years, or a rise of 4350 W degrees. The maximum for the 80-meter layer corresponds to an effective time of 18 years, or a rise of 12,100 W degrees. To be more specific, W = 0.01 watts/gal leads to a rise of 120°C for the 80-meter layer, 43°C for the 20-meter layer; if we put W = 0.04 in the 20-meter layer, the maximum rise is 174°C.

Attention should be called to the circumstance that it has been assumed, in effect, that the waste material is injected at the temperature of the rock of the reservoir, and that the equations are linear, so that solutions may be superposed. In particular, a linear normal rise of temperature with depth may be added to the solution found above. If the waste is hotter or cooler than the environment into which it is injected, there will be a correction term. This may be found for the conditions envisioned in the above examples. Suppose that the difference of initial temperature between the layer $-a<x<a$ and the surrounding rock is ΔT_o, uniform through the layer, and let this stay uniform within the layer throughout the process of cooling or heating to the surrounding temperature. Then the difference of temperature ΔT between the layer and the initial temperature of the surroundings is given by $\Delta T = \Delta T_o e^{z^2} \operatorname{erfc} z$, where $z = kn^2t/a^2$. This function is tabulated in Carslaw and Jaeger (p. 373). The points of interest are the times of maximum temperature rise resulting from the radioactive heating. For the 20-meter layer, this is about 50 years; this gives $\Delta T = 0.137\ \Delta T_o$. For the 80-meter layer, the maximum of heating comes at about 80 years, for which $\Delta T = 0.36\ \Delta T_o$. These corrections are unlikely to be large. Suppose, for example, that waste at 20°C is pumped into the pore space (20 percent) of rock at 120°C; the mixture will have an initial temperature of 87°C, so that ΔT_o is 33°. The corrections found above are thus less than 12° at the time of maximum rise from heating, in this case in the direction of reducing the maximum temperature. It would be injudicious indeed to work so close to the limit that this effect would be significant.

It has been assumed that the rise of temperature is uniform throughout the waste-filled layer. Another possible assumption would be that transport of heat

within the layer is by conduction alone, with the same thermal properties inside the layer as outside. The solution for steady heating at a constant rate A is given in Carslaw and Jaeger (1947, p. 61):

$$T = \frac{At}{c}\left\{1 - 2i^2\mathrm{erfc}\,\frac{a-x}{\sqrt{4kt}} - 2i^2\mathrm{erfc}\,\frac{a+x}{\sqrt{4kt}}\right\} \text{ for } 0<x<a\ ;$$

$$T \quad \frac{At}{c}\left\{2i^2\mathrm{erfc}\,\frac{x-a}{\sqrt{4kt}} - 2i^2\mathrm{erfc}\,\frac{x+a}{\sqrt{4kt}}\right\} \text{ for } x>a\ .$$

At the center,

$$x = 0,\ T(0) = \frac{At}{c}\left(1 - 4i^2\mathrm{erfc}\,\frac{a}{\sqrt{4kt}}\right);$$

at the edge,

$$x = a,\ T(a) = \frac{At}{2c}\left(1 - 4i^2\mathrm{erfc}\,\frac{2a}{\sqrt{4kt}}\right);$$

the center is always warmer than the edge of the layer, the rise of temperature being twice as great at the center as at the edge for short times, with the ratio approaching 1 for long times. For heating according to $A_o e^{-\alpha t}$, the corresponding solutions may be found to be

$$T(0) = \frac{A_o e^{-\alpha t}}{c}\int_0^t e^{\alpha\beta}\,\mathrm{erf}\,\frac{a}{\sqrt{4k\beta}}\,d\beta$$

and

$$T(a) = \frac{A_o e^{-\alpha t}}{2c}\int_0^t e^{\alpha\beta}\,\mathrm{erf}\,\frac{2a}{\sqrt{4k\beta}}\,d\beta\ .$$

Again, the center is always warmer than the edge, since erf x is always greater than (erf 2x)/ 2. These integrals not being at hand, we may estimate the difference for the case of steady heating, remembering that the absolute values so

obtained will be somewhat too high.[1/] With A/c = 6.7 deg/year, as assumed above (W = 0.01, porosity 20 percent) and an 80-meter layer, the rise in 20 years will be 119°C at the center, 67°C at the edge, a difference of 52°; as the radioactive decay in 20 years is to about 2/3 of the initial rate, the difference will be at least 35° when decay is allowed for. This means a gradient of 35/40 or about 1°/meter, and it is hardly likely that the layer would remain stable if the liquid were not sealed into the pore spaces (see below). The probable course of events seems to be that, supposing the layer itself to be bounded by impermeable formations, heating would at first take place as just described, with the center warming more than the boundaries. At some stage in this process, depending upon the permeability of the layer, instability would set in, and stirring would tend to bring the whole layer to a nearly uniform temperature. This process might be repeated a number of times. The long-term behavior is probably reasonably well described in terms of the heating of a layer of effectively infinite conductivity, for which the curves of Fig. 1 and Table 2 have been constructed.

Determination of Critical Conditions

The calculations of temperature for various assumed distributions of the heat sources are relatively straightforward. It is much more difficult to decide at what point the assumptions become physically unrealistic. Calculations leading to temperatures of thousands of degrees are obviously mere number-work; long before such temperatures were reached, the assumed conditions would be altered as new processes became important. The first question to be asked is, under what conditions will the liquid waste remain in a natural reservoir?

There is a great deal of structural and stratigraphic knowledge to be brought to bear from petroleum exploration and production but the likeness to a

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[1/] Dr. Walter Gautschi, National Bureau of Standards, points out that these integrals are expressible in terms of the complex error function. It can be shown that

$$T(0) = \frac{A_o e^{-\alpha t}}{c} \int_0^t e^{\alpha\beta} \operatorname{erf}\left(\frac{a}{\sqrt{4k}}\right) d\beta =$$

$$= \frac{A_o}{c\alpha}\left[\operatorname{erf}\left(\frac{a}{\sqrt{4kt}}\right) - e^{-\alpha t} + e^{-\frac{a^2}{4kt}} \operatorname{Rw}\left(\sqrt{\alpha t} + \frac{ai}{\sqrt{4kt}}\right)\right]$$

where

$$w(z) = e^{-z^2} \operatorname{erfc}(-iz) .$$

The function w(z) of the complex argument z is tabulated in V. N. Faddeeva and N. M. Terent'ev (1954).

petroleum trap is not so close as it may seem. It has been shown by Hubbert (1953) that capillary pressure amounting to at least several atmospheres aids in preventing the escape of oil or gas through a water-bearing fine-grained cap rock: "This phenomenon permits us to regard a sandshale boundary as an impermeable barrier to oil trapped in the sand, but not an impermeable barrier to the passage of water in either direction." Oil and gas move upward displacing water because of lower density and are trapped because immiscibility produces a barrier. Unlike oil or gas, aqueous wastes will be miscible with ground water and the density will be equal to or greater than that of ground water. It was for these reasons that the Committee on Waste Disposal recommended studying the possibility of disposing of liquid waste in porous and permeable sandstone strata in the deep parts of sedimentary basins. Those sandstones will be essentially horizontal and will be interbedded with other sedimentary rocks, all of which will be completely filled with ground water--probably high saline brines. The waste is or can be made denser than the water filling the pores and the waste should be placed in the lowest part of the structure from which it cannot migrate.

Layers of shale may have vanishingly low permeabilities, with self-sealing properties capable of closing up incipient fractures. Evaporites also form virtually impermeable layers, under natural conditions. In all of these cases, the native water is at the temperature of the surrounding rocks, with no exceptional internal heat generation. It seems likely that the heating of introduced liquid waste some hundred degrees or so would not produce drastic physical changes, and that plastic deformation of shale or evaporite layers, if sufficiently thick, might continue to provide a tight container. At some point, chemical or mineralogical changes must be expected; these, by altering the minerals, by solution effects, and so on, may open up channels through which the waste can move out of its supposed reservoir. These changes may be so slow that by the time they have a significant effect, the activity of the waste will have declined to a safe level; the Sr^{90} activity will be reduced by a factor of 1000 in 280 years.

It seems probable that in a reasonably permeable layer filled or partly filled with waste of the concentration we have been discussing (0.01 watts/gal) convection will be set up. The conditions for initiating convection in uniform permeable layers have been discussed by Lapwood (1948) and Goguel (1953). In a horizontal layer of uniform permeability σ, thickness h, the fluid is stable until the thermal gradient exceeds the adiabatic gradient by the amount β, given by $\beta = (4\pi^2 k\nu)/(h^2 \alpha g \sigma)$. Here α is the volumetric thermal expansion, ν the kinematic viscosity of the liquid, k the thermal diffusivity of the rock, g the acceleration of gravity. The critical thickness for $\beta = 30°/km$, of the order of the normal gradient, is shown in Table 3.

The critical thickness h is proportional to $1/\beta^{1/2}$; thus for gradients of the order of 1°/meter or 1000°/km, such as appeared in the discussion of the heated layer, the critical thickness would be roughly 1/6 as great as those of the table, other conditions remaining the same. For permeabilities of 1 darcy or greater, the critical thickness would be less than 40 meters, at 100°C.

TABLE 3

CRITICAL THICKNESS OF UNIFORM LAYER, FOR β = 30°/KM.

Permeability		Thickness, meters		
		Water		Steam
cm^2	darcys	20°C	100°C	250°C, 40 bars
10^{-10}	0.01	7260	2400	2100
10^{-9}	0.1	2300	750	660
10^{-8}	1	730	240	210
10^{-7}	10	230	75	66
Assumed parameters				
$10^4\alpha$, deg^{-1} =		2.5	7	30
ν, cm^2/sec =		0.01	0.003	0.009
k, cm^2/sec =		0.01	0.01	0.01

This is, of course, a high permeability, as rocks go, but something of this kind will probably be required to permit introduction of large quantities of waste. For 0.01 darcy, the critical thickness would be some 400 meters.

This circulation would be advantageous for the thermal problem, amounting indeed to a thickening of the layer; for a given amount of heat, we have seen that this reduces the maximum temperatures. Whether such a circulation would result in contamination of ground-water is another matter. Detailed studies of particular regions need to be made with all of the requirements in mind.

Another matter which needs some consideration is the possibility of boiling. The depth at which boiling becomes possible, for a given temperature, depends upon local conditions. It is usually found that the pressure in drill holes is somewhere between the pressure of a column of water to the depth in question and that of a column of rock of the same height. In thermal areas, the temperature-depth curves often come close to the curve which would be found by supposing that the water at every depth is at the saturation density. Saturation curves are shown in Fig. 2. The critical pressure is reached at 11,500 feet if the density is everywhere that of the saturated liquid, 7300 feet for unit density, 2900 feet for mean rock density of 2.5. Water at greater pressures would not boil; if, however, a channel developed by which the water could rapidly rise to higher levels, boiling might take place near the surface. This appears to be what happens in geyser

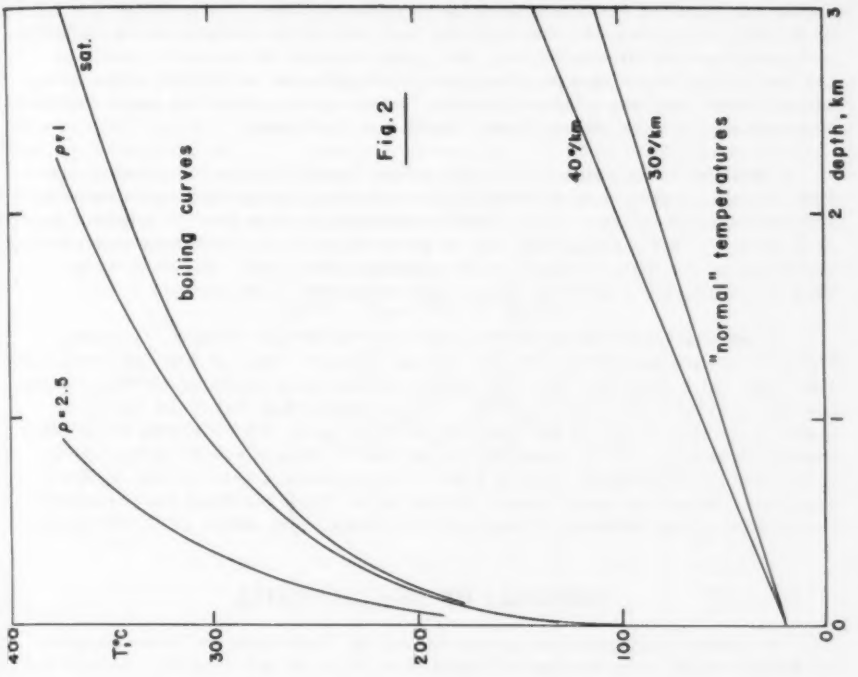
Fig. 2
T°C
400
300
200
100
0
sat.
ρ=1
ρ=2.5
boiling curves
40°/km
30°/km
"normal" temperatures
0
1
2
3
depth, km

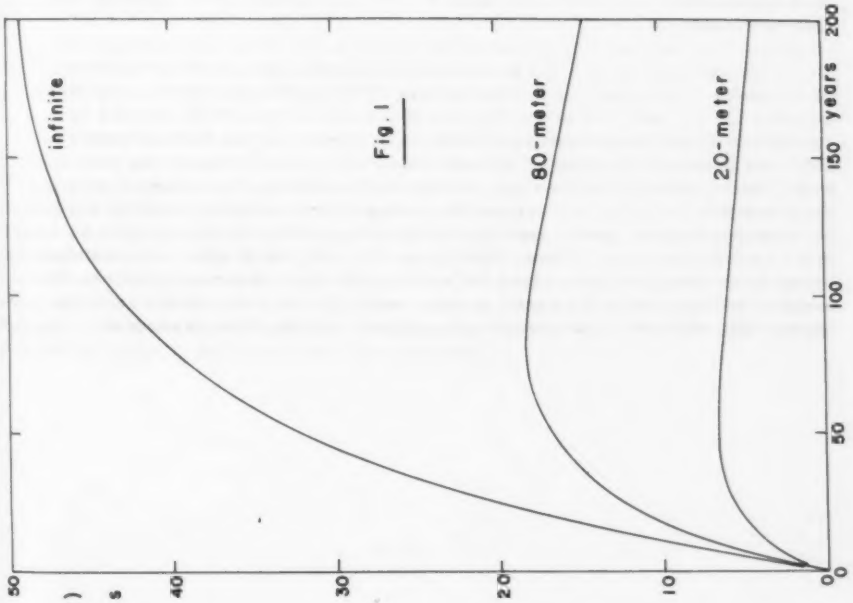
Fig. 1
f(t)
years
50
40
30
20
10
0
infinite
80-meter
20-meter
0
50
100
150
200
years

areas; at Wairakei, for example, large supplies of water at about 250°C are found at depths of the order of a few thousand feet, where the corresponding saturation temperatures are slightly higher. On moving upward, this water mixes with cooler surface water, and the temperature stays below the boiling point, except in the geysers and the artificial bores. The bores are producing some 1500 million gallons per year, as condensed steam and hot water.

The conditions which produced the New Zealand thermal area have persisted long enough to heat the near-surface rocks to temperatures close to the saturation temperatures. This takes a much longer time than the hundred years or so required for a single injection of waste to reach its maximum temperature, and escaping streams of waste would soon encounter cooler rock and water. This would provide a factor of safety against escape to the surface.

If we decide, somewhat arbitrarily, that the temperature of the waste should never exceed 300°C, then the deeper the injection, the smaller the allowable rise. At a depth of 2 km, the normal temperature might be 100°C or more, leaving a permissible rise of 200°C. This is about what was found for an 80-meter layer with W = 0.02 and porosity of 20 percent. There seems to be little reason, in general, for considering deeper burial, though specially favorable local features might lead to it. A 2-km layer, or even a 1-km layer, is thick enough to prevent any perceptible thermal effect from reaching the surface by conduction, unless the source continues for times of the order of 10,000 years.

DISPOSAL IN SALT DEPOSITS

A method of disposal recommended by the Committee on Waste Disposal for further study is by storage as liquid in cavities in salt deposits. One of the problems associated with this, as with other methods of storage, is to set safe limits with respect to the evolution of heat in the wastes. A tentative approach to this question may be based on the solution for the rise of temperature in a heat-generating layer, with allowance for the appropriate differences in the thermal constants.

The solution given for a heated layer depended upon time through two parameters, α the effective decay constant of the radioactive waste, and the quantity kn^2/a^2, where k is the thermal diffusivity (conductivity divided by specific heat) of the medium surrounding the layer, a is the half-thickness of the layer, and n is the ratio of specific heats c/c', c referring to the medium, and c' to the material of the layer itself. In the earlier discussion, it was supposed that the layer, as well as the surrounding medium, consisted of rock of about the same porosity, saturated with brine, and hence n was taken as 1, and k as 0.01 cm^2/sec. The solution is readily adapted to other assumptions, since, if we take different values for k and n, the time-dependence will be the same if we keep kn^2/a the same, in other words, change the thickness of the layer. The rise of temperature is proportional to the time-dependent

factor multiplied by A/c', A being the rate of heat generation per unit volume of the layer.

The thermal parameters of pure halite are reasonably well known, though there are some discordant values in the literature. Table 4 shows the conductivity for a natural single crystal (Birch and Clark, 1940), and the corresponding diffusivity, calculated with the specific heat as tabulated. The conductivity and diffusivity fall off as the temperature increases. The simple theory requires a diffusivity independent of temperature, and it will suffice for rough calculations to take k = 0.02 cm^2/sec, correct at about 120°C.

TABLE 4

THERMAL PARAMETERS OF HALITE

t, °C	K cal	c cal	k cm^2
	cm.sec.deg.	cm^3 deg.	sec.
0	0.0146	0.44	0.033
50	.0120	.45	.027
100	.0101	.45	.023
150	.0086	.46	.019
200	.0075	.47	.016
300	.0060	.49	.012
400	.0050	.50	.010

Tabular Cavities

We suppose that the layer is filled with brine, generating heat at the rate A = 2000 W cal/cm^3 year, where W is the rate in watts/gal. For the specific heat c', we take 1 cal/cm^3 deg. Thus A/c' will be 2000 W deg/year.

Compared with the former case, we now have n = 0.45, or say $n^2 = 0.2$, instead of 1, and k = 0.02 instead of 0.01. Thus kn^2 is now 0.004 instead of 0.010. To have the same time-dependence, a^2 must now be smaller, in the ratio 1/2.5, or a in the ratio 1/1.6. The curves given before for 20-meter and 80-meter layers now apply to 12.5-meter and 50-meter layers. These thinner layers contain five times as much liquid, per cm^3 as did the porous layers formerly treated. With respect to liquid capacity, the 20-meter porous layer was equivalent to only 4 meters, and the 80-meter porous layer to 16 meters. The better diffusivity of the salt has resulted in an improvement in the rate of dissipation by a factor of about 3, for this particular example.

We must, however, take into account the greater rate of heat generation. For the former 20-meter layer, we found that the maximum temperature was reached after about 50 years, and corresponded to heating with no loss at the initial rate for 6.5 years. This now applies to a liquid layer 12.5 meters thick in salt. The maximum temperature is then 6.5 x 2000 W degrees, or 13,000 W degrees. With W = 0.01 watt/gal, this is 130°C, etc. This is the rise of temperature in 50 years, to be added to the initial temperature. It is approximately the same rise that was found for the 80-meter layer with a porosity of 20 percent, the same concentration of waste, and k = 0.01 cm^2/sec. As this layer was equivalent to a layer of liquid 16 meters thick, the salt is less efficient than the porous layer, in the ratio 12.5 to 16 (78 percent).

The reason for this somewhat paradoxical conclusion is that in the porous layer, the thermal inertia, per unit volume of liquid, is greater than in the liquid itself, and by a factor which more than offsets the higher conductivity of the salt. The difference is not great enough to be decisive, however; we merely note that as between a layer of liquid in salt, and a layer in porous rock (with this order of porosity) there is no important thermal advantage, for the same amount of liquid. This leaves the decision to be determined on other grounds--accessibility, mechanical stability, effectiveness of sealing, etc. If a mined-out layer in a salt dome were used, there would be the advantage that the shape could be accurately surveyed, and the initial position of the liquid well determined, something which might be difficult in a porous structure. Enough pressure should be kept on the liquid to prevent boiling; this could most readily be accomplished with a liquid-filled standpipe reaching to the surface from an otherwise tight system. There should not be large vertical shafts which would permit convection and possible geysering. If the temperature were not allowed to rise above 150°C, the depth would not have to exceed about 150 feet.

Spherical Cavities

The solution for a uniform layer can evidently be used to give some information regarding the thermal effects of spherical cavities. Any number of such cavities of radius less than a can be distributed throughout a layer of thickness 2a, without exceeding anywhere the temperature found for the layer 2a, other parameters (A, porosity, thermal constants) remaining the same; the uniform layer gives the limit which is approached as the spheres tend to fill the whole layer. A single sphere, or widely spaced spheres, will have a diameter somewhat greater than the thickness of the uniform layer which would give the same temperature. The spherical cavity has some interest, moreover, as offering a possible experimental arrangement which would not require excessive times for reaching the maximum temperatures.

The calculation for spheres with exponential decay of the heating rate is straightforward, but the integrals required for numerical calculation are not at hand. For small spheres, a useful approximation may be obtained by

neglecting the decay, and computing the rise of temperature for a constant rate of heating. In this case, there is an ultimate steady-state temperature, different from zero. Let A/c be the rate of rise of temperature, if there is no loss of heat; the steady-state temperature outside the sphere is $Aa^3/3ckr$, and at the boundary, $r = a$, this is $Aa^2/3ck$. For a sphere of radius 10 meters, in a uniform conducting medium (the sphere also having the same conductivity) the temperature rise is 77 percent of the ultimate temperature in about 10 years, if $k = 0.02$ cm^2/sec, as for salt. Corresponding estimates for other sizes and times are shown in Table 5.

TABLE 5

TEMPERATURE RISE IN LIQUID-FILLED SPHERICAL CAVITIES IN SALT

Constant heating at 0.01 watts/gal.			
Radius, meters	Ultimate temperature °C	Time to reach fraction of ultimate temperature	
		77% years	50% years
5	2.6	2.45	0.4
10	10.4	9.8	1.6
20	41.6	39.2	6.2

The actual temperature rise depends upon the concentration. If this remains constant at 0.1 watts/gal, then the temperature rise in the sphere of 10-meter radius will be 80°C in 10 years. It will never much exceed this, the "steady-state" temperature rise being 104°C; in fact, because of the decay, the temperature after 10 years will be closer to 70°, and will thereafter rise only slightly to a maximum, then gradually decline. It is possible thus to control the maximum temperature and the time at which it will be reached by adjusting the concentration of the waste and the size of the cavity. The principal object of such an experiment would seem to be a large-scale verification of the thermal constants plus the opportunity to study other aspects of the interaction between solution and cavity. It could not provide a satisfactory test of the mechanical stability of much larger heated cavities in salt. The capacity of a sphere of 10-meter radius is about 4 million liters, roughly 1 million gallons. Evidently it is not essential that the cavity have an exact spherical form; the results will be much the same for any nearly equidimensional shape.

The "steady-state" temperature for spheres is proportional to Aa^2, while the capacity is proportional to Aa^3. For a given content of radioactive elements, consequently, the temperatures will be proportional to $1/a$ and it will be economical to use the largest possible sphere. This advantage is even

greater when we consider the transient case, as the larger the sphere the slower the rate of heating and the more important the effect of radioactive decay; this was also found to be true for the heated layer.

Solution For Heated, Conducting Sphere

The solution for the heated sphere in the uniform medium given in Carslaw and Jaeger (1947, p. 287-288) is incorrect. The problem is as follows: heat is produced for $t>0$ at the constant rate A per unit volume in the region $0\leq r<a$ of the infinite medium, of uniform conductivity K, diffusivity k, and zero initial temperature. The equations of heat conduction become,

for $0\leq r<a$,

$$c\frac{\partial T}{\partial t} = K\frac{\partial^2 T}{\partial r^2} + \frac{2}{r}\frac{\partial T}{\partial r} + A$$

and for $r>a$,

$$c\frac{\partial T}{\partial t} = K\left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r}\frac{\partial T}{\partial r}\right).$$

Applying the Laplace transformation, we obtain the subsidiary equations,

$$\frac{d^2(rv)}{dr^2} - q^2(rv) = -\frac{rA}{Kp}, \quad 0\leq r<a ;$$

$$= 0, \quad r>a .$$

The solutions are of the form,

$$rv = Ar/cp^2 + C \sinh qr, \quad 0\leq r<a ,$$

$$= B e^{-qr}, \quad r>a .$$

B and C are to be determined by the conditions that temperature and gradient must be continuous at $r = a$. Carslaw and Jaeger are correct to this point. The correct solutions for B and C are

$$B = (\cosh qa - \sinh qa/qa)\,(Aa/cp^2)$$

$$C = -(A/cp^2q)\,(1 + qa)e^{-qa}$$

Then for $0 \leq r < a$,

$$v = (A/cp^2) \left[1 - \sinh qr\, e^{-qa} (1 + qa)/qr,\right]$$

and for $r > a$,

$$v = (Aa/crp^2) e^{-qr} \left[\cosh qa - \sinh qa/qa\right].$$

The corresponding temperatures are

for $0 < r < a$,

$$T(r,t) = (At/c) \left[1 + \frac{2a}{r} \left(i^2\mathrm{erfc} \frac{a + r}{\sqrt{4kt}} - i^2\mathrm{erfc} \frac{a - r}{\sqrt{4kt}}\right)\right.$$

$$\left. + 2 \frac{\sqrt{4kt}}{r} \left(i^3\mathrm{erfc} \frac{a + r}{\sqrt{4kt}} - i^3\mathrm{erfc} \frac{a - r}{\sqrt{4kt}}\right)\right]$$

and for $r > a$,

$$T(r,t) = (2Aat/cr) \left[i^2\mathrm{erfc} \frac{r - a}{\sqrt{4kt}} + i^2\mathrm{erfc} \frac{r + a}{\sqrt{4kt}}\right.$$

$$\left. + \frac{\sqrt{4kt}}{a} \left(i^3\mathrm{erfc} \frac{r + a}{\sqrt{4kt}} - i^3\mathrm{erfc} \frac{r - a}{\sqrt{4kt}}\right)\right].$$

Temperatures may readily be calculated for any values of r and t with the aid of the tabulations of $i^2\mathrm{erfc}\, x$ and $i^3\mathrm{erfc}\, x$ in Carslaw and Jaeger (1947). The steady-state temperatures are,

for $0 \leq r < a$,

$$T(r) = (Aa^2/2ck) (1 - r^2/3a^2) ;$$

and for $r > a$,

$$T(r) = Aa^3/3ckr$$

This value for $r > a$, in the steady-state, is independent of the distribution of the heat sources in the sphere of radius a, so long as point symmetry is preserved, the total quantity Aa^3 remaining the same.

The solution for the same problem but with exponential decay of the heating rate is obtained from the foregoing solution by Duhamel's theorem.

For r = o, the solution is

$$T(0,t) = \frac{A_o a^2}{2ck} \left\{ e^{-\alpha t} \int_{a/\sqrt{4kt}}^{\infty} e^{\alpha a^2/4k\beta^2} \left(\operatorname{erf} \alpha - \frac{2\beta}{\sqrt{\pi}} e^{-\beta^2} \right) \frac{d\beta}{\beta^3} \right\}$$

for r = a,

$$T(a,t) = \frac{A_o a^2}{3ck} \left\{ 3e^{-\alpha t} \int_{a/\sqrt{kt}}^{\infty} e^{\alpha a^2/k\beta^2} \left(\operatorname{erf} \beta - \frac{2}{\beta\sqrt{\pi}} (1 - e^{-\beta^2}) \right) \frac{d\beta}{\beta^3} \right\}$$

These may be written, for given α,k,a,

$$T_o(t) = (A_o a^2/2ck)\ F_o(t)\ ,$$

for r = 0,

$$T_a(t) = (A_o a^2/3ck)\ F_a(t)\ .$$

The terms in parentheses are the steady-state temperatures which would be reached for constant heating at the rate A_o(no decay); the functions $F_o(t)$ and $F_a(t)$ give the fractions of these temperatures reached at the time t and r = 0 and r = a, respectively. Numerical results are shown in Table 6 for r = a, α = 0.02 per year, k = 0.02 $cm^2/sec.$ = $64.10^4 cm^2/year$, a = 10 meters. For comparison, the corresponding figures for steady heating are also shown.

This particular solution does not correspond to the case of a liquid waste, since the sphere r = a is filled with salt, exactly like the surrounding material except that heat is supposed to be generated in it. Such a situation might arise if the radioactive elements were distributed in a matrix of dry salt, which was then packed into a spherical cavity. The figures given above will be too high for the case of a cavity in salt filled with liquid waste, though the difference should not be great for spheres of small radius (say, a = 10 meters or less).

Waste-Filled Spherical Cavity in Salt

A closer approach to the problem of a cavity in salt, filled with liquid waste, may be obtained as follows: suppose the cavity to be spherical, of

TABLE 6

FRACTION OF TEMPERATURE $Aa^2/3ck$
REACHED IN t YEARS,
a = 10 meters, k = 0.02 cm^2/sec

t years	Heating according to	
	$A\ e^{-\alpha t}$ α = 0.02/year percent	A (constant) percent
0	0	0
1.6	49	50
6.3	66	
9.8	66	77
17.4	62	
39.1	44	87
61.0	30	
108.5	13	
156.3	6	
infinite	0	100

radius a, completely filled with liquid of specific heat, per cm^3, c', and heat production A, per cm^3. Suppose that the temperature is uniform throughout the sphere, because of convection within the liquid, and that conduction takes place into an infinite mass of salt. The initial temperature is zero throughout. The boundary condition is

$$\left.\frac{\partial T}{\partial t}\right|_a = \frac{A}{c'} + \frac{3K}{c'a}\left.\frac{\partial T}{\partial r}\right|_a$$

Applying the the Laplace transform, and taking the solution of the subsidiary equation, for r>a, as $\bar{v} = B\ e^{-qr}/r$, we find the solution in the form

$$\bar{v} = \frac{Aa}{c'kr}\ \frac{e^{-q(r-a)}}{p(q+\alpha)(q+\beta)}$$

where $-\alpha$ and $-\beta$, are the roots of $(q^2 + \mu q/a + \mu/a^2)$. As before, $\mu = 3\dot{c}/c'$, three times the ratio of (volume) specific heats. For the case of interest, μ is about 1.35, so that α,β are complex. We have $\alpha,\beta = (\mu/2a)\ (1 - i\sqrt{4/\mu - 1})$.

The solution is then, r>a,

$$T(r,t) = \frac{Aa}{c'kr} \frac{1}{2\pi i} \int_{\gamma-i\infty}^{\gamma+i\infty} \frac{e^{-q(r-a)+pt}}{p(q+\alpha)(q+\beta)} dp .$$

On integration, the temperature is found to be

$$T(r,t) = \frac{Aa^3}{3ckr} \left\{ 1 - \frac{2\mu}{\pi a^2(\beta-\alpha)} \int_0^\infty \left[\frac{\cos \sigma x + \alpha \sin \sigma x / x}{x^2+\alpha^2} - \frac{\cos \sigma x + \beta \sin \sigma x / x}{x^2+\beta^2} \right] e^{-ktx^2} dx \right\} .$$

Here $\sigma = r - a$. We need only the temperature of the solution, at r = a or $\sigma = 0$. This is

$$T(a,t) = \frac{Aa^2}{3ck} \left\{ 1 - \frac{2\mu}{\pi a^2(\beta-\alpha)} \int_0^\infty \left[\frac{1}{x^2+\alpha^2} - \frac{1}{x^2+\beta^2} \right] e^{-ktx^2} dx \right\}$$

$$= \frac{Aa^2}{3ck} \left\{ 1 - \frac{2}{\pi} \int_0^\infty \frac{e^{-kty^2/a^2} dy}{(y^2/\mu - 1)^2 + y^2} \right\} \quad \text{footnote 2/} .$$

In this form, we have only to evaluate a real, definite integral; for small values of a, the exponential factor quickly approaches zero, so that numerical integration is convenient.

2/ Dr. Walter Gautschi, National Bureau of Standards, points out that this integral can be expressed in terms of the function w(z). Namely,

$$\frac{2}{\pi} \int_0^\infty \frac{e^{-kty^2/a^2}}{(\frac{y^2}{\mu} - 1)^2 + y^2} = \sqrt{\frac{kt}{a^2}} \frac{\mu}{1 - \frac{\mu}{4}} I \left\{ \frac{w(z)}{-iz} \right\}$$

$$z = \sqrt{\frac{kt}{a^2}} \left[\sqrt{\mu(1 - \frac{\mu}{4})} + i \frac{\mu}{2} \right] .$$

Reference: Faddeeva and Terent'ev, *op.cit.*

For heating according to $A_o e^{-\alpha t}$, we apply Duhamel's theorem, and obtain the solution for r = a, in the form of a definite integral,

$$T(a,t) = \frac{2A_o\, e^{-\alpha t}}{3\pi c} \int_0^\infty \frac{[\,1 - e^{-(ky^2/a^2 - \alpha)t}\,]\, y^2\, dy}{(ky^2/a^2 - \alpha)\,[\,(y^2/\mu - 1)^2 + y^2\,]} .$$

This is also relatively convenient for numerical computation. A few results are given below for a = 10 meters, α = 0.02 per year, k = 0.02 cm^2/sec = 64.10^4 $cm^2/year$. The temperatures at time t are given by A_o/c' multiplied by a factor having the dimensions of time (years); this factor may be called the effective heating time $\bar{t}$. We may also express the results as fractions of the fictitious steady state temperature which would be reached if the rate of heat production remained constant and equal to A_o. As can be seen above, this is $A_o a^2/3ck$. These percentages may be compared directly with the results given above for the case of the conducting sphere in which heat is generated. There is a slight lag with respect to the earlier results as a consequence of the greater heat capacity of the solution, but the difference for a 10-meter radius is slight.

TABLE 7

RESULTS FOR WASTE-FILLED SPHERE, OF 10-METER RADIUS, IN SALT

t years	$(1 - e^{-\alpha t})/\alpha$ years	$\bar{t}$ years	$\frac{\alpha\bar{t}}{(1 - e^{-\alpha t})}$ percent	Factor of $A_o a^2/3ck$ percent
7.81	7.23	0.76	10.5	66
15.63	13.42	0.73	5.4	63
31.25	23.24	0.59	2.5	51

It is also useful to calculate the fraction of total heat generated within the sphere which is retained in the waste. For a constant rate of heating, there is an asymptotic approach to a limiting temperature, even though the total heat input increases in proportion to the time. Thus the fraction of heat retained approaches zero in the limit. The same thing is true for exponential decay of the heating rate, except that now the total heat input is finite. The amount generated in time t is proportional to $(1 - e^{-\alpha t})/\alpha$, approaching a limit, for α = 0.02 per year, of 50 years. This time is also shown in Table 7; comparison with $\bar{t}$ which is proportional to the temperature rise at time t, shows that after 7.81 years, for example, close to the time of maximum rise, only 10.5 percent of the integrated heat input is retained in the waste. Thus the sphere of 10-meter radius, in salt, is a fairly effective dissipating system. The maximum temperature rise is about $0.8\, A_o/c'$ = (0.8) 2000 W, where as before W is the initial heating rate in watts/gallon. For W = 0.1, the maximum rise would be about 160°C.

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Summary Report

REACTOR FUEL WASTE DISPOSAL PROJECT

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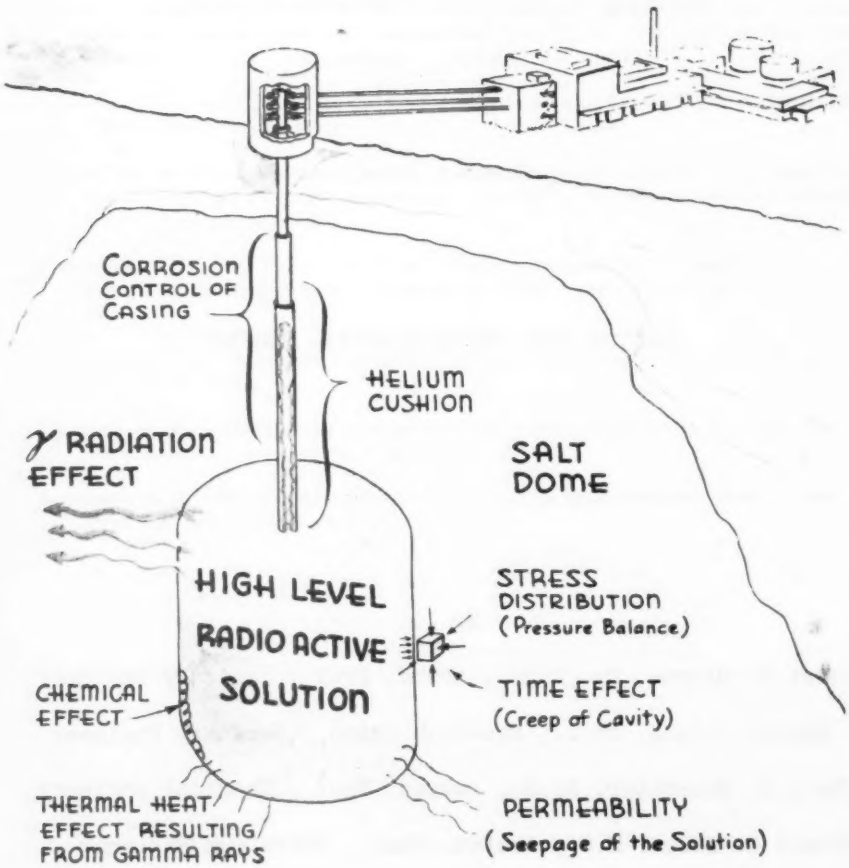
REACTOR FUEL STORAGE PROBLEM

RESEARCH CONCEPTS FOR STORING WASTE



THE DISPOSAL OF HIGH-LEVEL RADIOACTIVE WASTES INTO SALT FORMATIONS

Abstract

This paper summarizes research performed at The University of Texas on the storage of reactor fuel wastes in salt formations. A few of the problems investigated include: (1) geological studies of salt domes, (2) structural stability, (3) permeability, (4) thermal heat effect, (5) creep, (6) stress distribution, (7) time effect, (8) chemical effect, (9) radiation effect, (10) cost of developing solution cavities, and (11) development of design principle for disposal into salt cavities.

Theoretical analyses based on model tests and limited field studies have demonstrated that a salt cavity can be designed such that it is structurally stable. Although thermal factors and plastic properties of salt are very important considerations, sufficient evidence now exists which will permit the design of storage structures.

The average cost to complete an average sized cavity in a salt dome for liquefied petroleum gas storage is $1.05 per barrel of storage space.

Introduction

Hundreds of millions of gallons of liquid petroleum products have been successfully stored in salt cavities by oil companies and the petrochemical industry.[1,2,3] Approximately 75 per cent of the total storage of liquefied petroleum gas products has been in either salt beds or salt domes (39.3 and 35.8 per cents, respectively).[4] A large cavity in a salt deposit can be constructed with much less expense than any other type of storage tank. The present rate of expense for storage

tanks is $0.25 to $2.00 per gallon of constructed cavity space. Many salt cavities of 10 to 100 million gallon capacity have been constructed with a capital investment of a few cents per stored gallon. If the mined-out cavity in a salt dome can be used successfully, the unit storage expense will be further reduced. The total space created by mining rock salt during a period of twenty years, 1934 to 1953, in the nation, was calculated to be about seven billion gallons[5].

Geology of Salt Domes and Storage Possibilities

Salt beds and domes occur in abundance throughout the country[6], in an area of more than half a million miles square, extending through eighteen states, as shown in Fig. 1. These deposits, which are located along the south side of the Great Lakes and along the Gulf Coast, have been considered most seriously for the disposal of radioactive waste because of their convenient location for industries. The Great Lakes deposits range in thickness from 200 to 2,000 feet, at depths of 500 to 10,000 feet. The depth of many Gulf Coast salt domes is less than a few thousand feet, making it feasible to construct underground storage cavities economically.

Since the geology of bedded salt deposits has been discussed by others[7], only the salt domes and their possible use as ultimate disposal sites is discussed in this section.

Distribution of Salt Domes

Intrusive masses of salt are widely distributed throughout the world. They are found in North America, Columbia, Germany, Roumania, France, Poland, Spain, Iran, Arabia, Palestine, U.S.S.R., Morocco, Tunisia, and Algeria. In North America, salt domes are present in the United States and Mexico. Those

in the United States are found in Texas, Louisiana, Arkansas, Mississippi, Colorado, and Utah, Fig. 1.

The largest group of salt domes in the United States, of the 200 known groups, lies adjacent to the Gulf of Mexico in southeast Texas and southern Louisiana. There is also a small group of salt domes in south Texas near the Rio Grande. An interior belt of salt domes in the Gulf Coast region is located in eastern Texas, northern Louisiana, southern Arkansas, and Mississippi. The reader is referred to Fig. 2 for a more exact location of the salt domes of the Gulf Coast Province. The salt of these structures is believed to be of Permian and/or Jurassic (Louann) age. The position of this salt layer is shown in Figs. 3, 4, 5, 6, and 7. The relative thickness of the Louann salt is seen in Fig. 6. The geologic units recognized in the Gulf Coast Province are graphically illustrated in Fig. 7.

Several salt anticlines and salt domes are recognized in southeastern Utah and southwestern Colorado. The salt in these deposits is thought to be of Pennsylvanian age. In this region salt reaches the surface at several salt structures.

Geology of Salt Domes

Some Gulf Coast salt domes are expressed topographically as hills, which rise a few feet to 40 feet above the surrounding lowlands, and in exceptional instances, as much as 200 feet. These topographic highs may be a mile or more in diameter. Depressions above some of the salt domes may be occupied by lakes and/or swamps.

The salt masses in the domes of the Gulf Coastal Plain, in plan view, are typically circular or broadly elliptical. Their average diameter is approximately 2 miles although they may range in diameter from 0.5 miles to 4 miles or more.

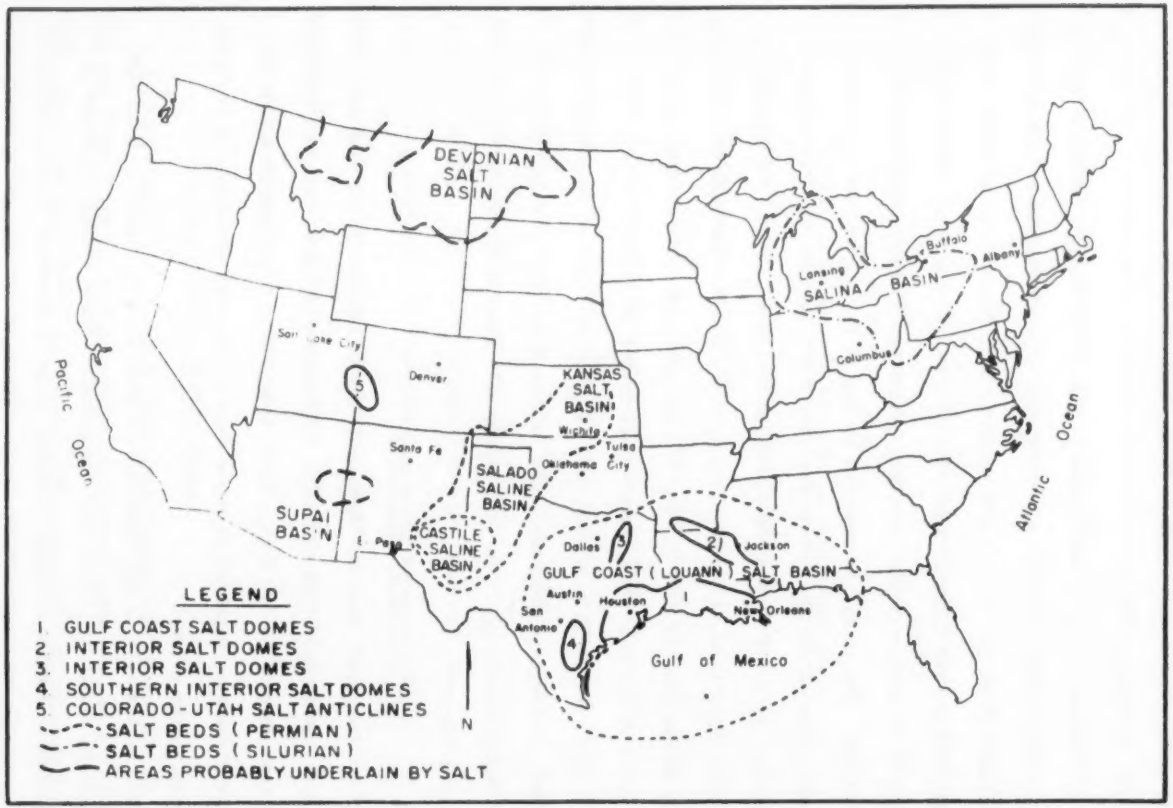


FIG. 1 PRIMARY SALINE DEPOSITS IN THE UNITED STATES.

FIG. 1 PRIMARY SALINE DEPOSITS IN THE UNITED STATES.

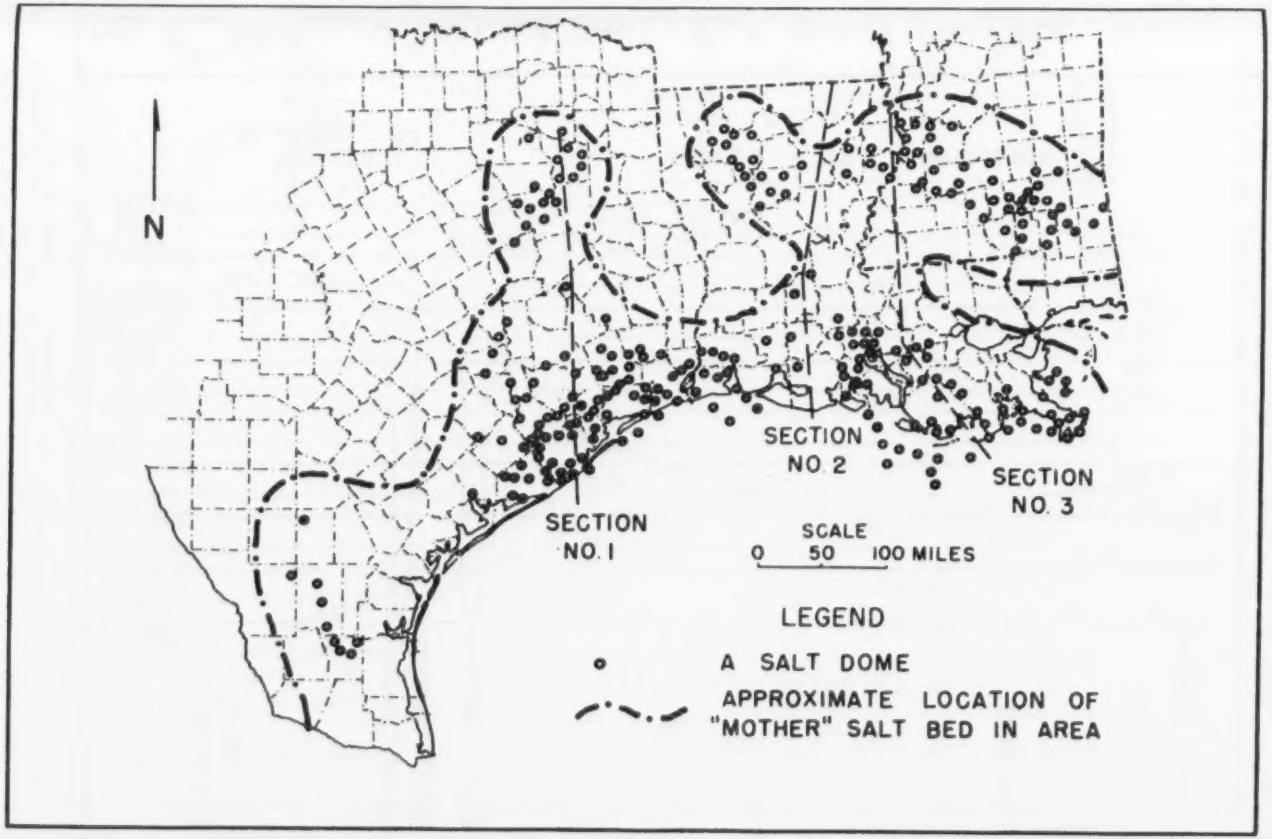


FIG. 2 MAP OF GULF COAST PROVINCE SALT DOMES

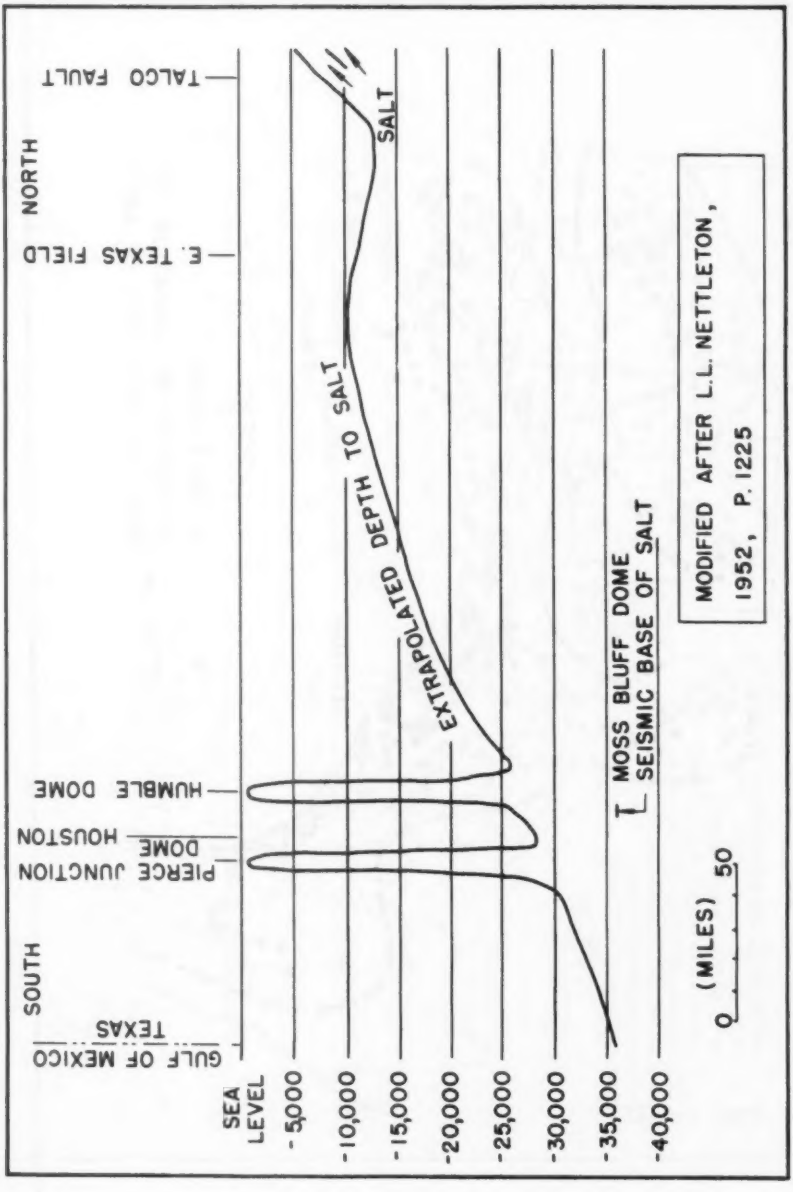


FIG. 3 DEPTH TO "MOTHER" SALT, GULF COAST PROVINCE - SECTION I

FIG. 3 DEPTH TO "MOTHER" SALT, GULF COAST PROVINCE - SECTION 1

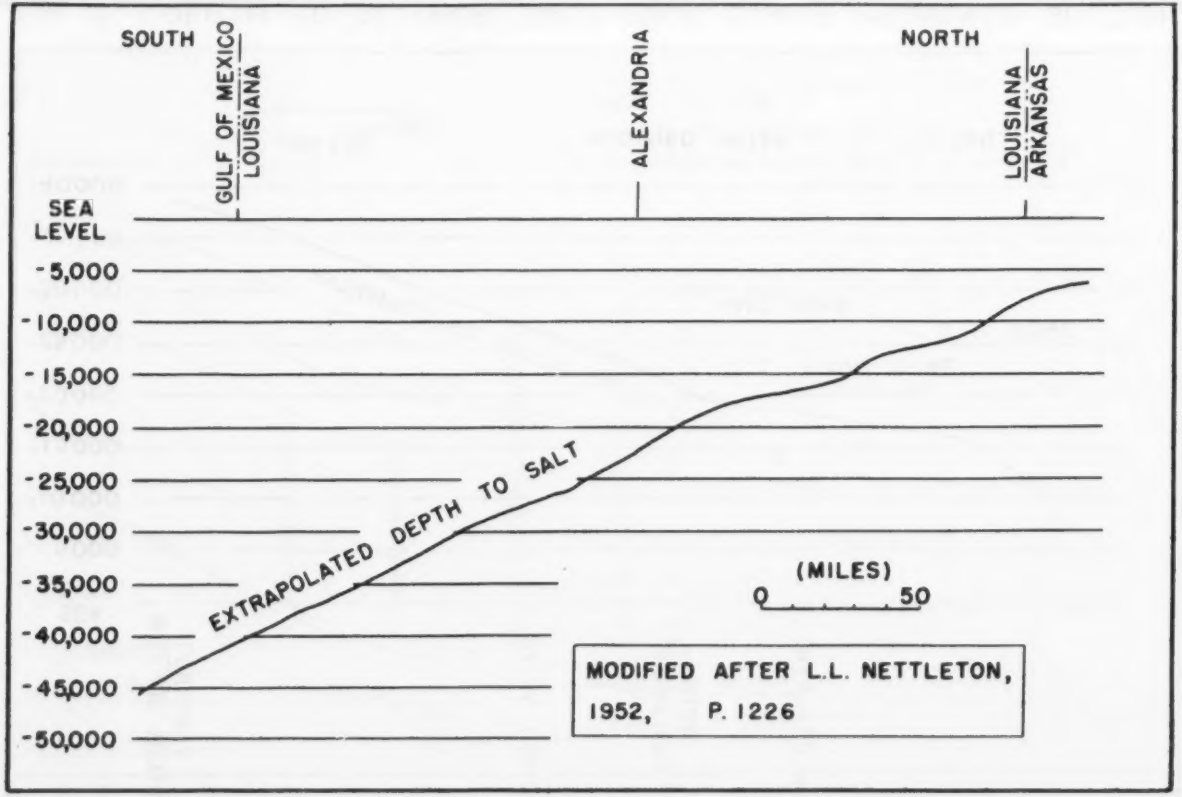


FIG. 4 DEPTH TO "MOTHER" SALT, GULF COAST PROVINCE - SECTION 2

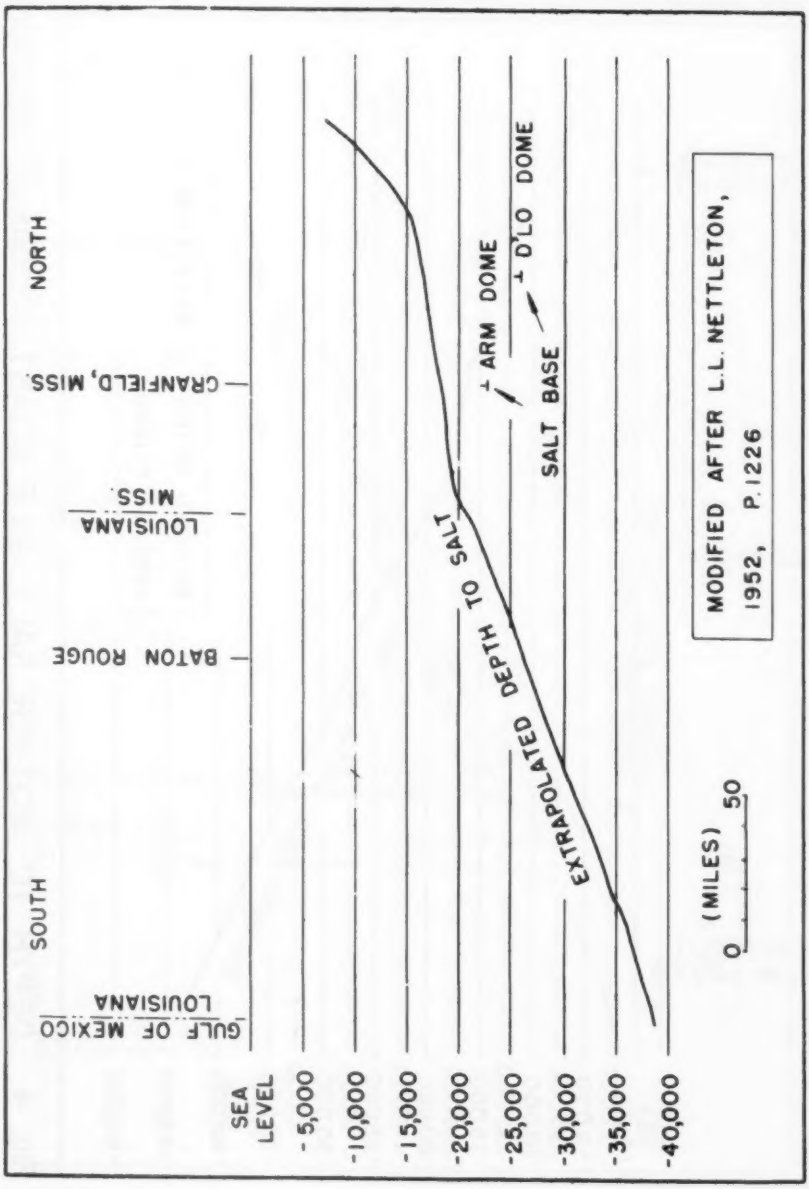


FIG. 5 DEPTH TO "MOTHER" SALT, GULF COAST PROVINCE - SECTION 3

FIG. 5 DEPTH TO "MOTHER" SALT, GULF COAST PROVINCE - SECTION 3

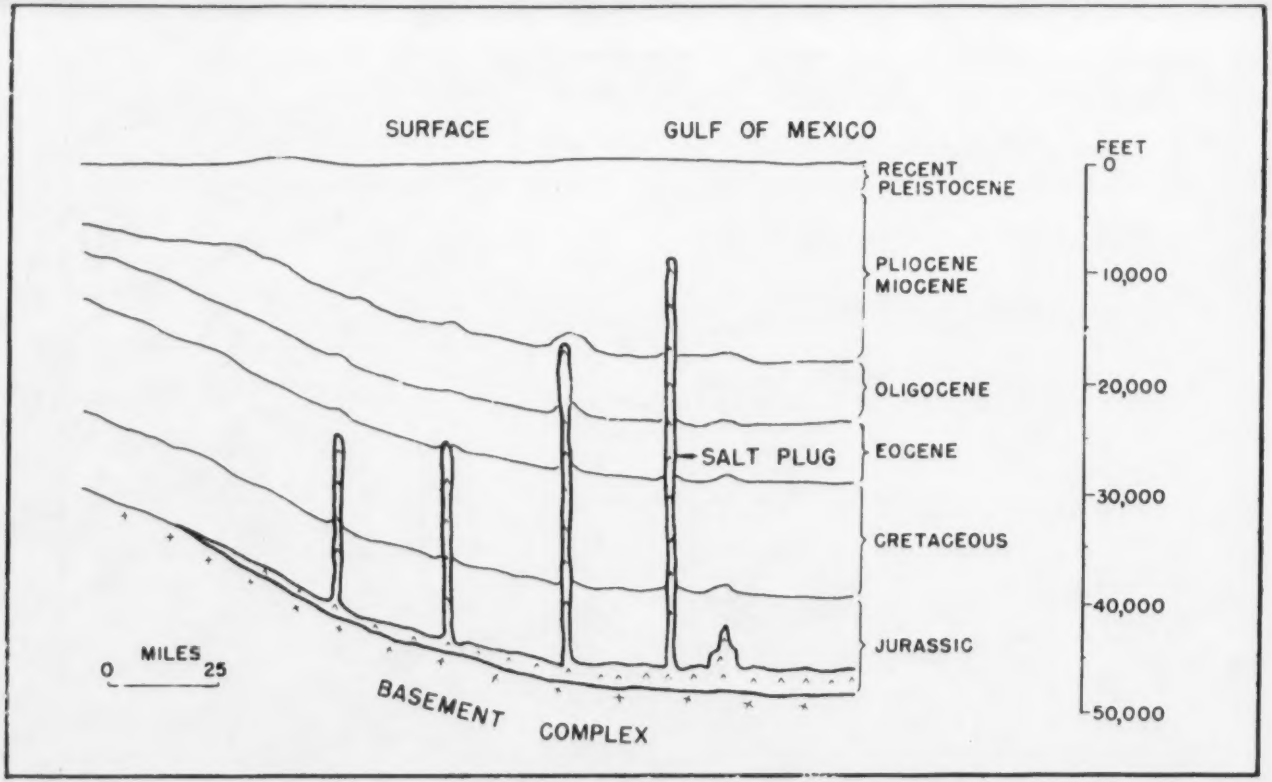


FIG. 6 GENERALIZED SECTION SHOWING THE NATURE OF THE FORMATIONS WITH SALT DOMES OF THE GULF COAST PROVINCE

SYSTEM	SERIES	GROUP or STAGE	THICKNESS
CENOZOIC — QUATERNARY	RECENT		105,400 +
	PLEISTOCENE		
CENOZOIC — TERTIARY	PLIOCENE		
	MIOCENE	GRAND GULF	
	OLIGOCENE	VICKSBURG	
	EOCENE	JACKSONIAN	
		CLAIBORNE	
		WILCOX	
	PALEOCENE	MIDWAY	
MESOZOIC — CRETACEOUS	GULFIAN	NAVARRO	99,350 +
		TAYLOR	
		AUSTIN	
		EAGLEFORD	
		WOODBINE (TUSCALOOSA)	
	COMANCHEAN	WASHITA	
		FREDERICKSBURG	
		TRINITY (INCLUDING TRAVIS PEAK OF TEXAS)	
	COAHUILAN	NUEVO LEON & DURANGO GROUPS OF MEXICO (HOSSTON-SLIGO)	
MESOZOIC — JURASSIC		COTTON VALLEY	60,525 +
		HAYNESVILLE	
		SMACKOVER } LOUARK GROUP	
		NORPHLET	
MESOZOIC — PRE-JURASSIC (?)		LOUANN	
PALEOZOIC PRECAMBIAN — BASEMENT COMPLEX			

MODIFIED AFTER G. E. MURRAY, 1952, P. 1157

FIG. 7 GENERALIZED GEOLOGIC COLUMN
CENTRAL GULF COASTAL PLAIN

Some of the salt masses are nearly cylindrical, but downward enlargement of diameter is common so that most of the domes have the form of a truncated cone. In some cones the upper part of the salt overhangs on one or more sides in a mushroom-like development. The upper surface of the salt in most domes is flat or slightly convex upward, but on a few domes irregular or even sharply pointed masses of salt project above the general level. The salt mass is sheathed by one or more layers of shale, gouge, or anhydrite, depending on the number of faults parallel to its surface.

The strata above the salt of the deeper domes are arched upward and dip away from the center of the affected area, but beds which have been pierced by the salt are turned up sharply, and in some domes stand almost vertically against the salt. The structural closure on some horizons adjacent to the salt domes amounts to several thousands of feet. (If a formation is entirely enclosed by one or more elevation contours, it is known as a closed structure.) In general, the closure is greatest on deeper horizons and decreases upward so that no deformation is observed at the surface and near-surface strata over some deep domes.

Normal faults are numerous in the strata above and adjacent to the salt mass, but thrust faults and reverse faults appear to be rare. Most of the salt plugs of the Gulf Coast have no faults, thus presenting a large, circular, unfractured section of salt.

Although the salt of caprock of some salt domes occurs at or very near the surface, the tops of other domes lie at depths of many thousands of feet.

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On some uplifts, however, salt has not been encountered even by very deep drilling, although geophysical data indicate its presence at a great depth. In general, domes that occur at depths of less than 4,000 feet are classed as shallow, those between 4,000 and 10,000 feet as intermediate, and those at still greater depths are referred to as deep.

The massive salt plugs developed along the Gulf Coastal Plain, as seen in Fig. 6, have moved up through the overlying rocks for distances as much as 30,000 feet. They are, in fact, still moving in terms of geological time as is shown by their surface expression as either mounds or depressions in the topography of the Recent surface of the Coastal Plain. The source of the salt is believed to be in southward extensions of the Jurassic and Permian salt formations that are found by drilling farther up the regional dip, Figs. 6 and 7.

Nearly all the salt domes of the Gulf Coast Province have a disk-like plate called the cap rock. This cap rock is usually composed of anhydrite, gypsum, limestone, and/or dolomitic limestone, with comparatively minor amounts of sulphur and other minerals. Most cap rocks are found to be both highly porous and permeable.

Deformation of Salt

Under stress, halite crystals deform by slip on the six dodecahedral planes (110, Miller indices), in the direction $(1\bar{1}0)$.[8] The least shearing stress that will cause permanent deformation of unconfined halite crystals is 853 psi.[9] There is agreement that dry, impure halite is stronger than dry, pure halite; that enclosed water increases its strength greatly; that previously strained salt is stronger than unstrained salt; and that salt

FIGURE 8. Effects of plastic flow of salt on 4 inch by 4 inch by 8 foot timber. The timber is located 400 feet southeast of main shaft, and has been in place about 10 years.

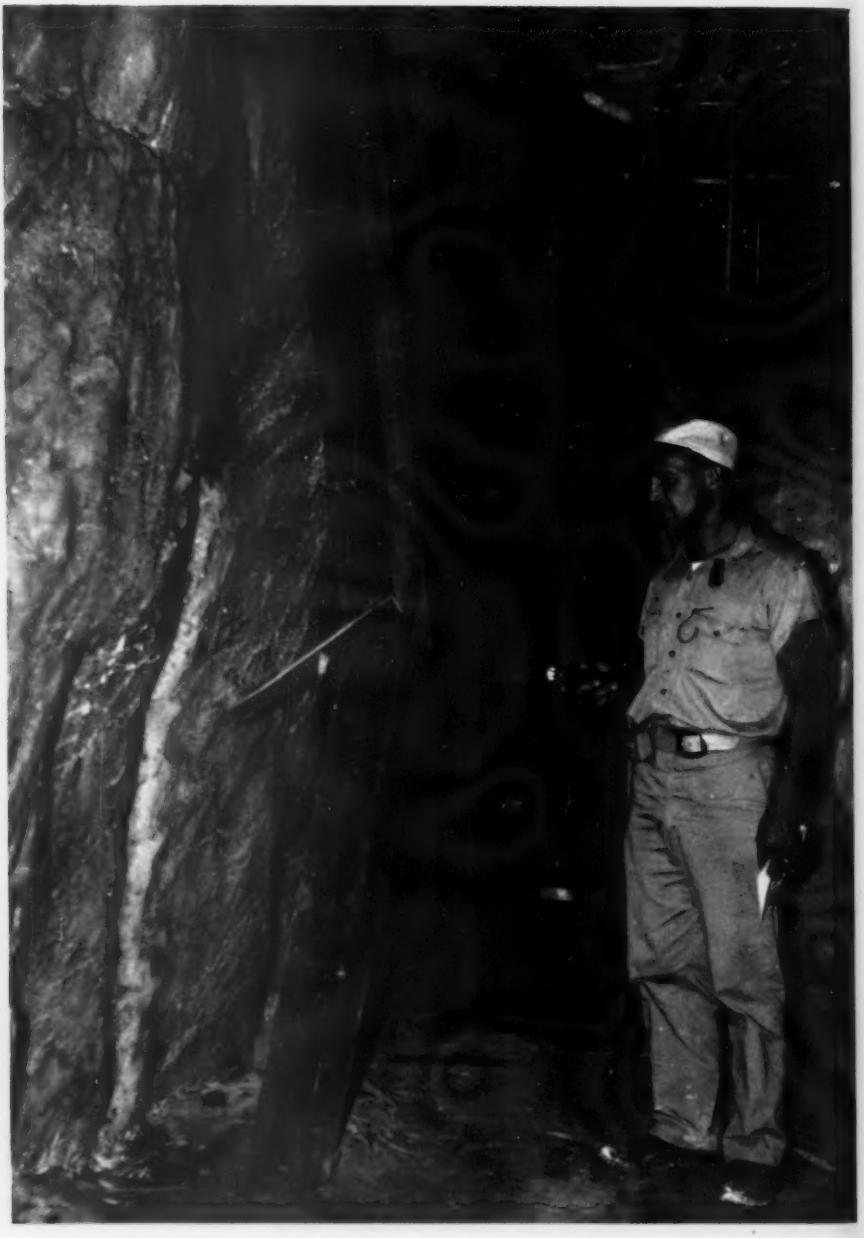

FIGURE 9. Typical failure resulting from plastic flow.

FIGURE 10. Shear folding caused by slip along planes dipping to the right in photo. Dark layers are anhydrite-rich salt. Height of face about 25 feet.

FIGURE 11. Nearly vertical parallel banding exposed on west wall of room. Light colored, nearly horizontal wavy streaks are caused by salt dust. Wall length about 50 feet.

at low temperatures is stronger than at high temperatures. Whether an aggregate of small halite crystals is stronger, as strong, or weaker than a single crystal, does not yet seem to be clear.[8]

From the above, it is apparent that there is a rather clearly defined stress at which a given salt crystal will begin to flow at an appreciable rate. Below this stress the rate of strain is so slow, that it requires long-time tests to measure it. In this range, the flow is called creep.[19] Creep in halite had not been tested extensively in the laboratory until recently, but a few observations have been made in salt mines. Figs. 8 and 9 are examples of the effect of creep.

Features of the Salt

The most obvious feature of the salt seen at the Grand Saline salt dome in Texas is the layering caused by bands of salt which contain a higher concentration of small (0.1 mm) anhydrite crystals, as in Fig. 10. These crystals are separate entities surrounded by the salt. The fact that they absorb more light than the NaCl makes these anhydrite bands stand out as dark gray layers. The darker the layer, the higher the concentration of anhydrite. The darkest bands may contain as much as 15 percent anhydrite. These alternating bands of pure salt and anhydrite-rich salt are believed to be beds now deformed from their original horizontal depositional position, see Fig. 11. All interpretations of the structure of salt are based on this belief. Undeformed salt also shows layering of this type.

This layering has been folded, and is very impressive when observed in the ceiling of the mine. The folds have been tipped on end by the upwelling of the salt into the dome, so that the walls show only the nearly vertical streaks and the contortions of the folds are seen best

in the ceilings. The map shows the position of the anhydrite-rich beds and demonstrates their intricate folding. When traced southwest from near the eastern boundary, the major folds become more attenuated and more compressed. The zig-zag pattern observed on the axial portions of the fold is more difficult to observe progressively southwest, but it appears to increase in amplitude. These zig-zags are shear folds caused by plastic failure of the salt along narrow closely-spaced bands.

Thermal Considerations

The storage of high-level radioactive wastes introduces a problem which is not encountered in the underground storage of petroleum products. The energy released by the radioactive wastes in the form of ionizing radiation is converted into heat. Because of practical difficulties, it is unlikely that this heat generated as a result of radioactive decay will be removed from the cavity by an external sink. The thermal energy must be dissipated to the environment; consequently, the temperature within the cavity will rise above that of the surroundings.

The underground storage of high-level wastes is inviting only if it provides containment both safe and economical. There are many aspects to be considered in satisfying the latter requirement. The fact that salt is more plastic at higher temperatures and that the waste is a source of energy is not in itself a reason for rejecting the idea of underground storage since this energy can be diluted. It is necessary, however, to know what the temperature rise might be within a cavity as a function of the possible parameters.

The rate of heat generation of a radioactive waste is a function of

the specific power of the reactor in which the fission products were generated, the time of irradiation in the reactor, and the time lapsed since discharged from the reactor. The heat generation per unit volume of waste is also a function of the concentration of fission products in the process effluent. The level of concentration is taken to be 800 gallons of waste per ton of uranium processed in all calculations except where specially noted.

Figures 12 and 13 demonstrate the magnitude of the temperature increase for a waste which was generated in a reactor operated at a power level of 33 1/3 megawatts per ton of uranium and irradiated to a total of 3,300 megawatt days per ton. This particular waste has been precooled 6 years prior to storage. Fig. 12 indicates the possible temperature rise within cavities of different radii as a function of time. Fig. 13 shows the spatial distribution of temperatures as a function of time.

Note that the parameter t as shown in Fig. 13 is a dimensionless time and the actual time for a particular cavity can be found by multiplying by the radius of the cavity squared and dividing by the thermal diffusivity.

As was indicated above, it may be desirable to limit the temperature increase by diluting the waste material prior to storage. The dilution (volume of water per volume of waste) required to limit the temperature rise to less than 100° F or 200° F is given in Fig. 14 as a function of the radius of the storage cavity.

As a result of defining the temperature limits for a particular waste it is now possible to evaluate the structural problem.

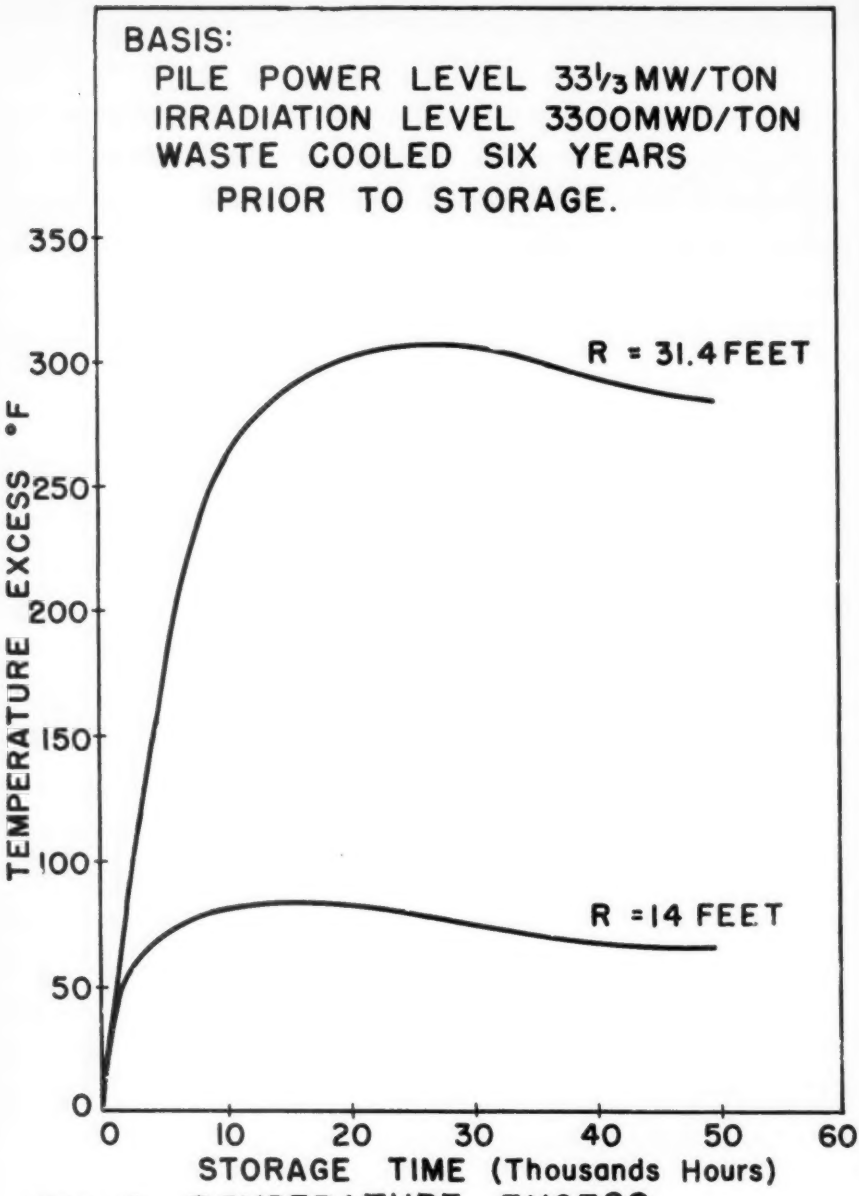

FIG. 12 TEMPERATURE EXCESS

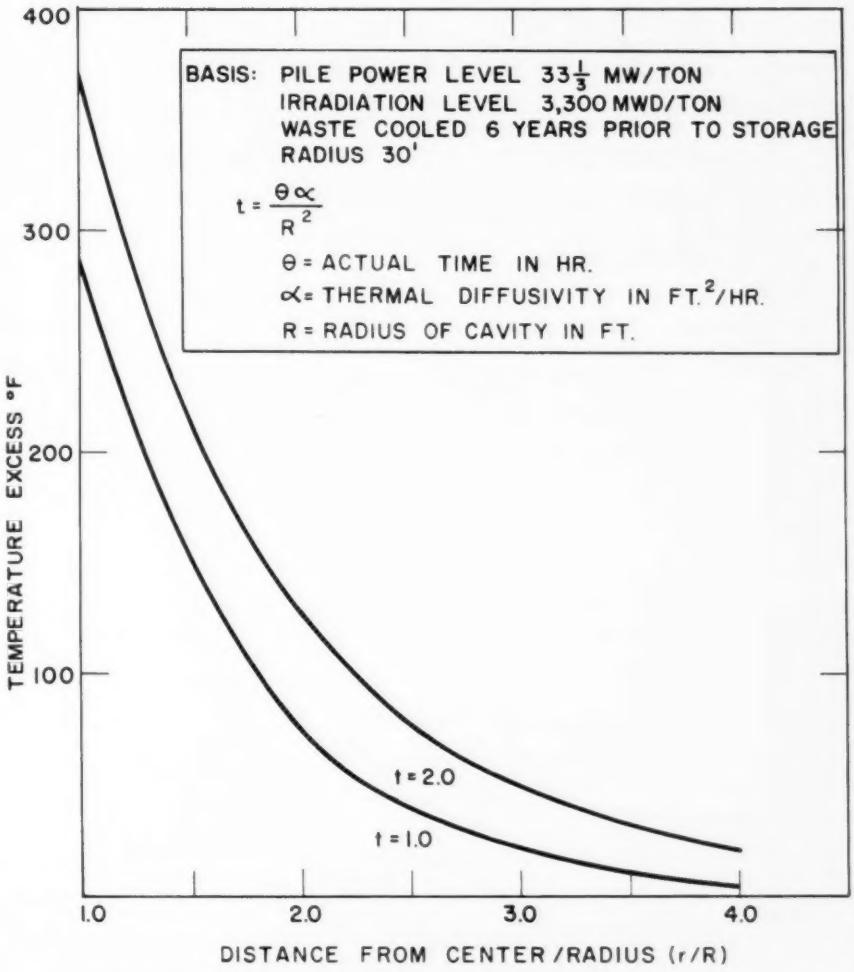

FIGURE 1.3 TEMPERATURE EXCESS SHOWN AS A FUNCTION OF TIME AND POSITION

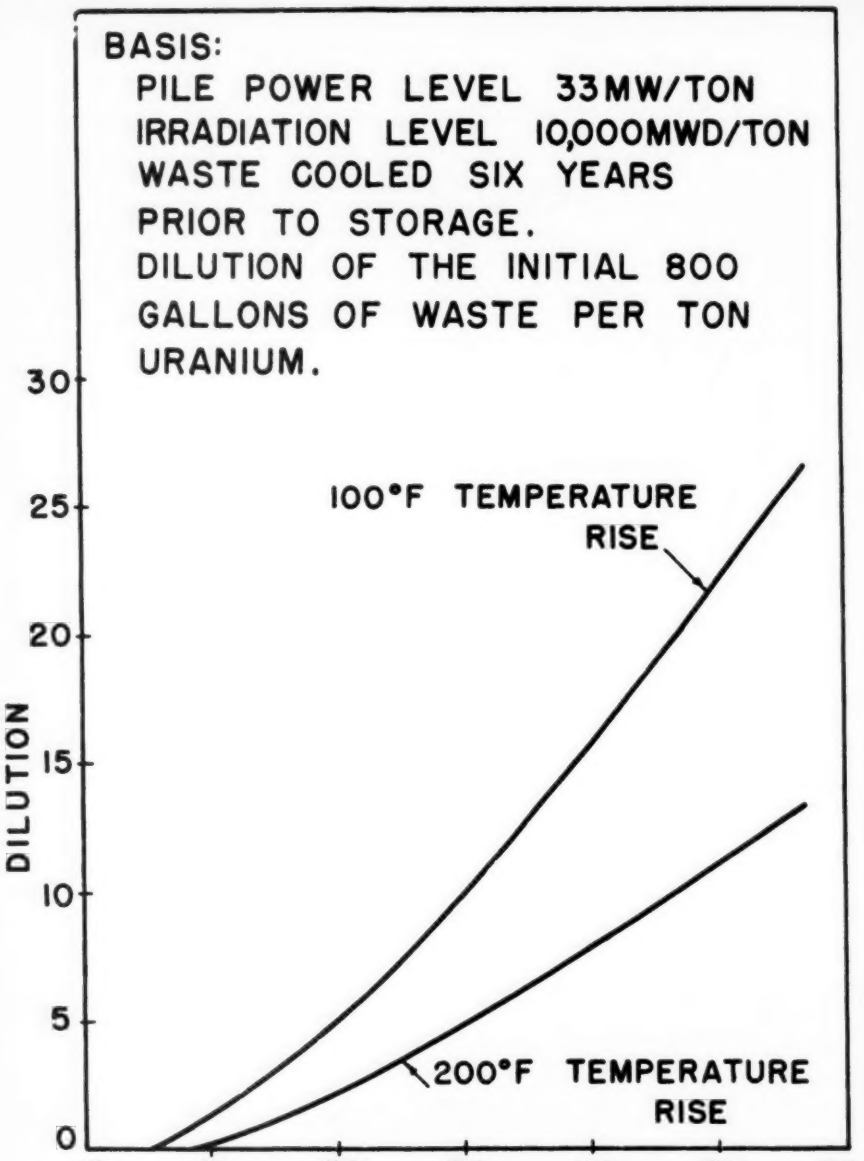

FIGURE 14 DILUTION REQUIREMENTS

Structural Stability

Although extensive experience has been acquired in the mining of salt, very little study has been made on the structural stability of salt cavities, particularly in the case of elevated temperatures. This is because salt cavities and tunnels at the present mining depth of a few thousand feet are strong enough without need for additional support. Hence, no need has arisen for structural investigation of a salt cavity. However, studies on the structural behavior of salt have been conducted by many investigators.[12-20] Aside from these reports, most of the previous publications about salt have been geological surveys and tectonic explanations rather than structural studies. No reference has been found on stress analysis of salt cavity, or on the strength of salt structures.

Elastic Stress Analyses

Stress acting around a cavity opening in a solid medium is the principal cause of either plastic deformation or collapse of the cavity. Since stress is not a property, but a state of material, the stress around a cavity can be mathematically determined, regardless of the nature of the material of which the cavity is made.

The theories of elasticity and photoelasticity can be applied to stress analysis of underground salt cavities within a certain depth, depending upon cavity geometry. For theoretical analysis of the elastic stress, it is reasonable to assume that salt formations (both salt domes and salt beds) are homogeneous and isotropic. The size of the salt formation is assumed to have infinite dimensions compared with the size of the cavity opening.

Furthermore, any additional lateral pressure acting on the salt formation has been assumed to be static and less significant than the pressure due to

overburden load at the depth of cavity opening. In general, regions in which salt deposits occur are effectively at rest in terms of hundreds of years.

Thus, the major stress existing in a salt formation to be considered is that due to the overburden load. However, if any additional lateral pressure is known to be acting on the salt formation, the stress distribution around the cavity due to this lateral pressure can be obtained by the same analysis as that of the overburden load, and then the one superimposed over the other, as illustrated in Fig. 15.

Effect of Depth of the Overburden Layer

The effect of depth of the overburden layer above a cavity opening upon the stress concentration around the opening has attracted much attention from civil engineers because it is a most important structural problem in the construction of tunnel openings. The maximum stress at ceiling, floor and rib of a circular horizontal tunnel of infinite length is shown in Figs. 16 and 17 as a function of the ratio of overburden depth to opening diameter. In the diagram, the stresses are expressed in terms of the stress concentration factors, that is, the value of stress expressed by overburden pressure as a measuring unit. A plus sign of the factor indicates tensile stress and a minus sign indicates compressive stress. It is apparent that the effect of the overburden depth on the stress concentration factor diminishes very rapidly with an increase in depth, and the overburden effect becomes insignificant at a depth of more than three times the diameter of the opening; beyond which the stress concentration factor comes close to the constant value. A comparison of the three groups of different initial stress conditions indicates that the initial stress condition of a formation is a controlling factor of the stress distribution around an opening at any depth.

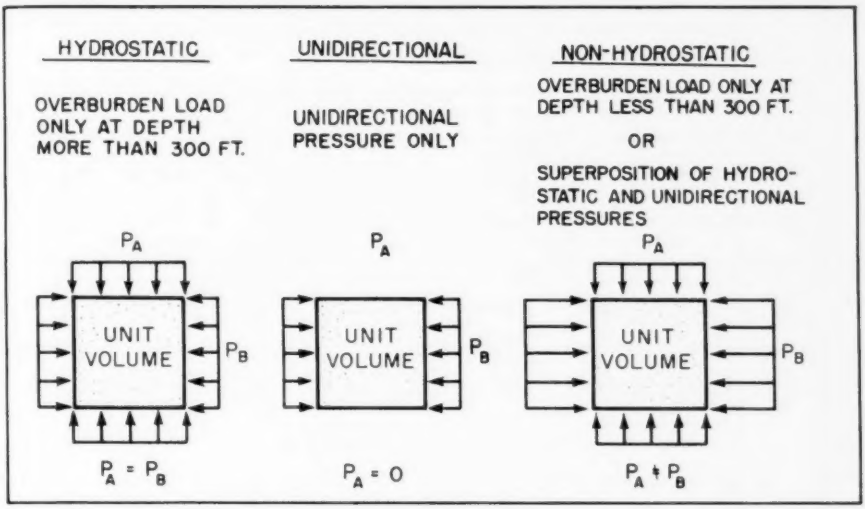

FIG. 15 INITIAL STRESS CONDITIONS IN SALT FORMATION.

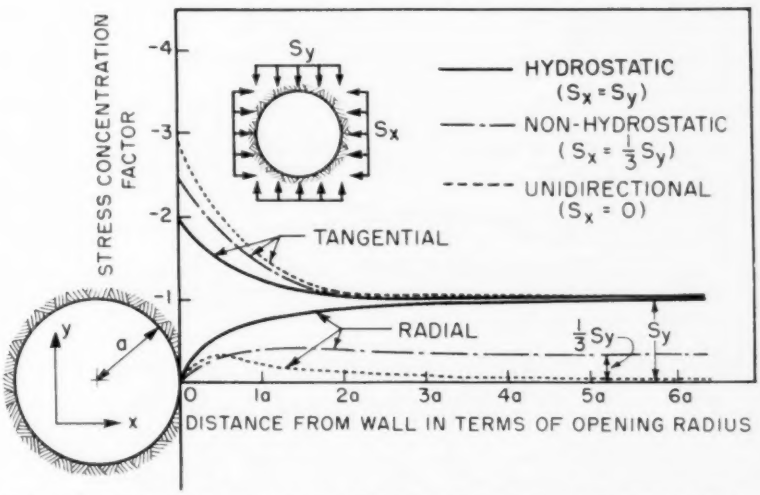

FIG 16 STRESS DISTRIBUTION ALONG THE HORIZONTAL DIRECTION FROM THE BOUNDARY OF CIRCULAR TUNNEL OPENING.

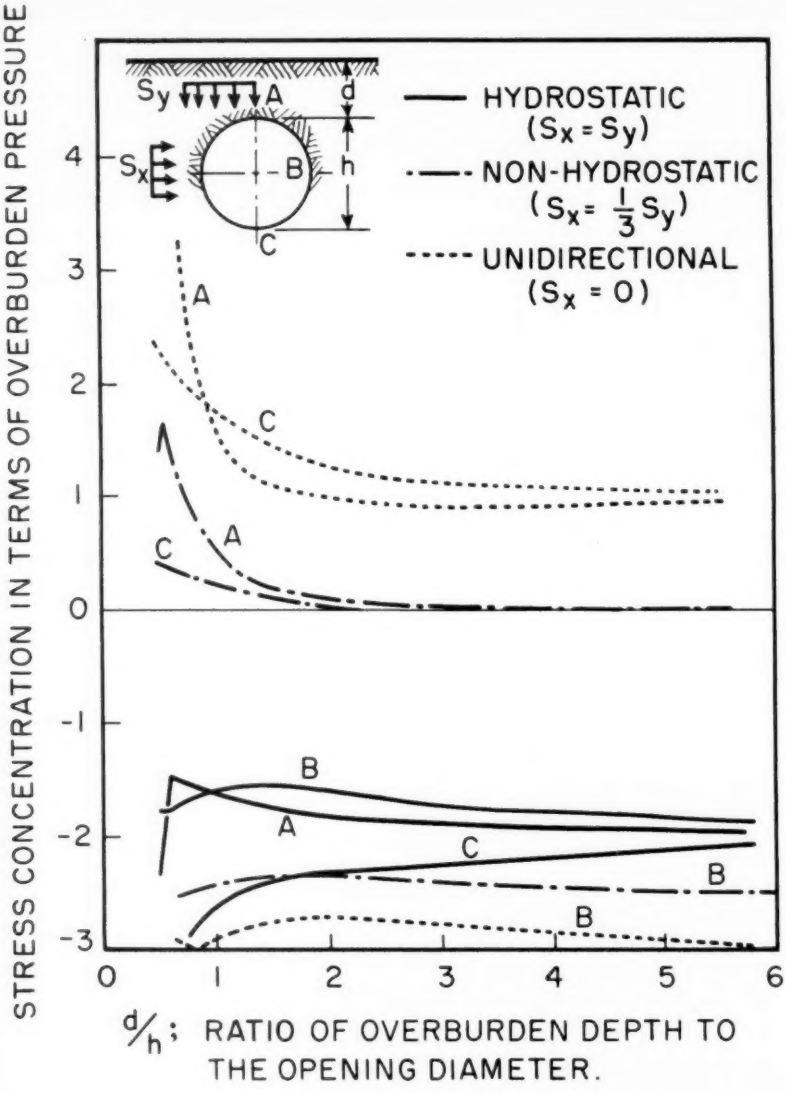

FIG. 17 EFFECT OF DEPTH OF OVERBURDEN LAYER ON TANGENTIAL STRESS CONCENTRATION AROUND THE BOUNDARY OF CIRCULAR TUNNEL OPENING.

In the hydrostatic condition, probably in most salt formations at a depth of more than 300 feet, no tensile stress occurs around the opening, and the stress on the floor (at C) first becomes critical at a shallow depth. At a depth less than the opening diameter, the stress concentration factor rapidly increases on both ceiling and floor; and the cavity becomes very unstable. Thus, the larger the size of cavity the larger the depth of the overburden layer should be,in order to have the same stress concentration factor. There should be however, a depth at which stress becomes minimal because the deeper the cavity, the greater the overburden pressure, and the less the stress concentration factor. Therefore, the depth should be a few times that of the opening diameter.

Initially Existing Stress in Salt Formations

The initial stress naturally existing in any underground formation before an opening is made is primarily determined by two factors: the overburden load and the Poisson's ratio of the formation.

The relation between the principal stresses and the Poisson's ratio can be derived mathematically from the fundamental stress-strain formulas in three dimensions. The strains in an element in underground formation on three mutually perpendicular directions (x, y, and z) can be described as:

$$\epsilon_x = \frac{1}{E}\left[S_x - \nu\,(S_y + S_z)\right]$$

$$\epsilon_y = \frac{1}{E}\left[S_y - \nu(S_z + S_x)\right] \qquad (1)$$

$$\epsilon_z = \frac{1}{E}\left[S_z - \nu(S_z + S_y)\right]$$

where

e_x, ϵ_y = strains in horizontal position

ϵ_z = strain in vertical direction

S_x, S_y = stress in horizontal direction

S_z = stress in vertical direction

E = Young's modulus

ν = Poisson's ratio

Since no lateral expansion of the element due to the overburden load is possible in an underground formation, and the vertical stress is equal to the overburden load, the following relations can be substituted in Eq. 1:

$$S_z = \text{vertical stress} = \text{overburden load}$$

$$S_x = S_y = \text{lateral pressure}$$

$$\epsilon_x = \epsilon_y = 0$$

The result is:

$$S_y = \frac{\nu}{1 - \nu} S_z \qquad (2)$$

Therefore, this equation means that the vertical stress, S_z, in an underground formation is always equal to its overburden pressure, and the lateral stresses, S_x and S_y, have greater values than zero and equal to or less than the overburden load, depending upon the Poisson's ratio of the formation.

To determine the initially existing stress distributions in salt, experimental application of the Poisson's ratio is necessary. Results of the laboratory experiments as well as calculations on seismic data agree that the value of the Poisson's ratio is approximately 0.5 at depths of 300 feet or more, which makes the stress distribution in the salt hydrostatic.

Effect of Size of Cavity Opening

Both mathematical analysis of stress and experimental observation by photoelasticity have proven that, in an initially stressed medium, the extent of the influence of cavity creation upon stress distribution is directly proportional to the size of the opening. The maximum stress always appears on the boundary of the cavity in a tangential direction, the minimum stress appearing on the boundary in a radial direction.

However, the stresses are independent of the size of the cavity and constant at a certain depth of the cavity.

Stress Concentration around Various Forms of Opening

Maximum and minimum stresses and maximum shearing stress always appear on the boundary around a cavity opening; also, the magnitude of these critical stresses is determined by the geometry of the cavity opening. The distribution of critical stress around the boundary depends mainly upon two factors, the initial stress condition of the formation and the form of cavity opening.

As seen in Figs. 18 and 19, the high concentration of shearing stress, due to opening a cavity in salt, is limited to close proximity of the opening. There is no shearing stress when the internal pressure of a cavity, P_1, is made equal to the overburden pressure of the salt formation, P_o. While the distance from the opening increases, the maximum shearing stress, Q_{max}, decreases much faster in a spherical cavity than in the long cylindrical cavity.

The problem of stress concentration around a number of openings created in close proximity has been solved by both mathematical and experimental methods; The latter being solved by using photoelasticity techniques. Thus, strees distribution around any form of cavity made in a salt formation can be determined provided the maximum shearing stress does not exceed the octahedral shearing strength of salt.

Plastic Analyses

If a salt cavity is to be used for storage disposal of reactor fuel wastes, plasticity of salt is probably the most important influential factor in structural stability of the cavity. The concentrated stress

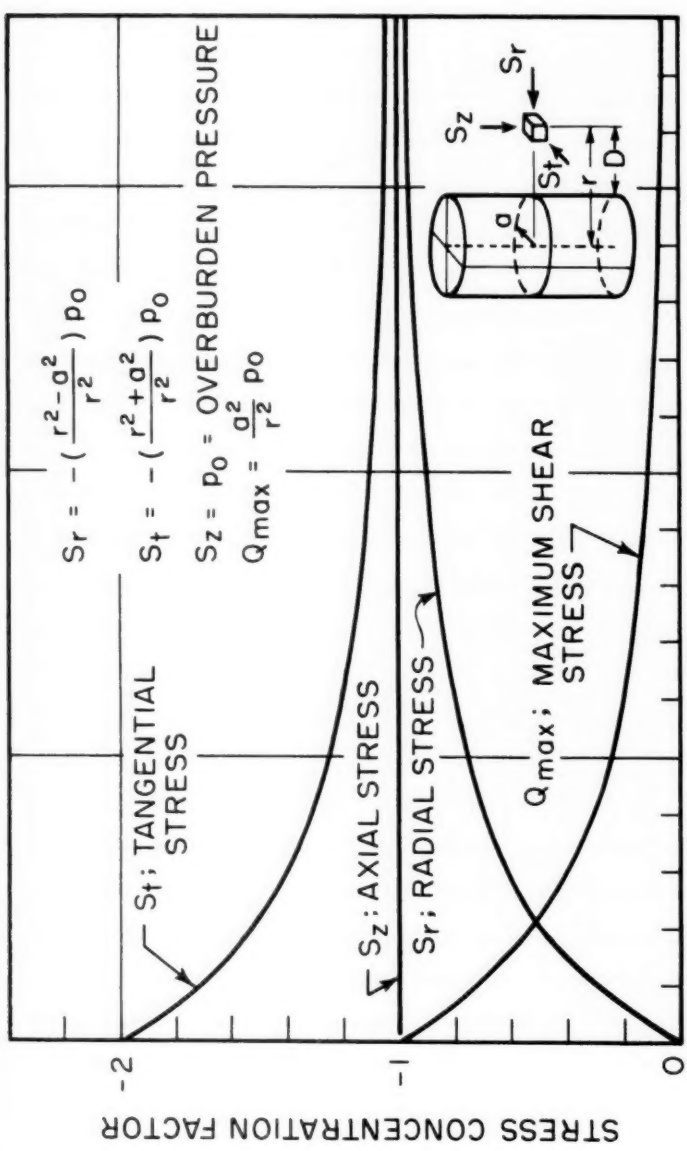

FIG. 18 STRESS DISTRIBUTION AROUND LONG CYLINDRICAL CAVITY.

CYLINDRICAL CAVITY.

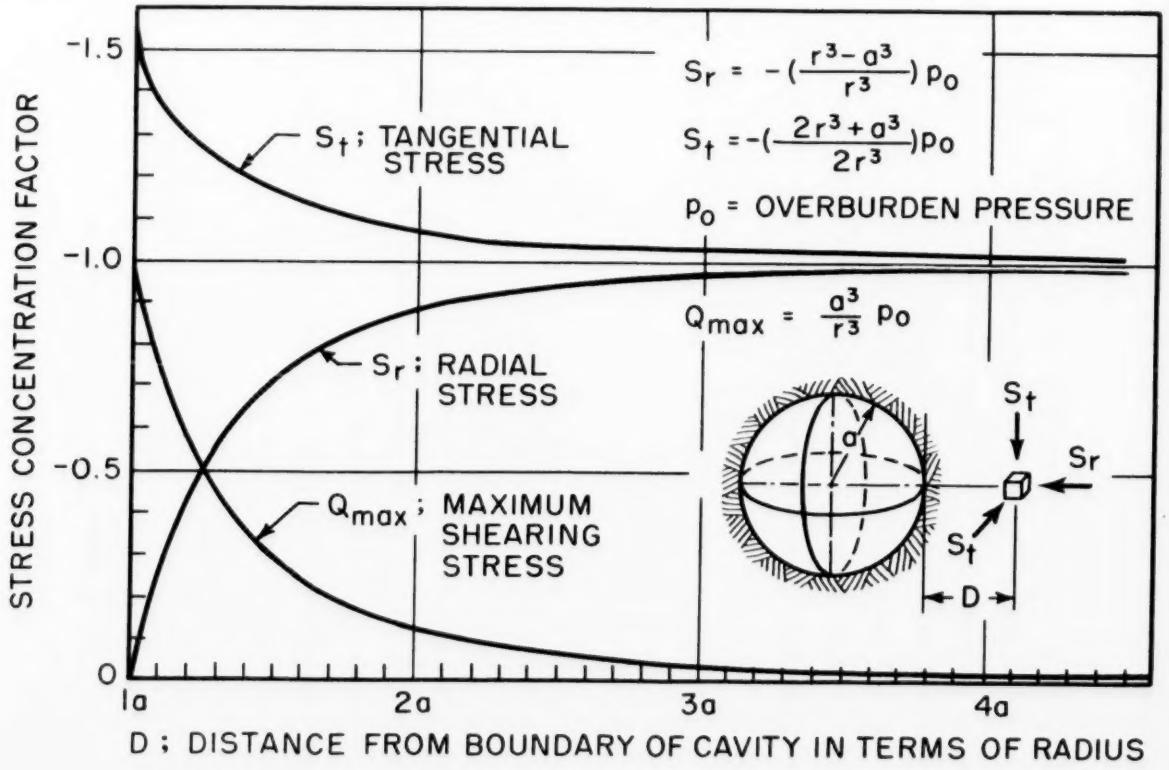

FIG. 19 STRESS DISTRIBUTION AROUND SPHERICAL CAVITY

resulting from creation of the cavity is reduced to a certain tolerable value by plastic deformation of salt around the cavity. This plastic deformation starts when the difference between the external and internal pressures acting on the salt cavity exceeds a certain value yet the salt cavity will never collapse under an increased pressure difference. It will merely widen the plastic zone surrounding the cavity. Thermal stress produces a further extension of the plastic zone.

From the analysis on the ideal forms of a salt cavity, it is clear that the thickness of the plastic zone depends entirely on the structural loading factor, X, which is defined as:

$$X = \frac{P_o - P_i}{\sigma_o}$$

where P_o = overburden load of cavity

P_i = internal pressure of cavity

σ_o = equivalent plastic limit of salt

And the increase of the radius of the plastic zone, ρ, can be obtained as an exponential function of this factor X. The stress distribution in plastic and elastic zones around the cavities at various depths is shown in Figs. 20 and 21. In conclusion of the plasticity analysis, a salt cavity can withstand unlimited structural loading by extending the size of its plastic zone, if the plastic zone can be extended to adjacent formations. However, the structural loading is limited by the following restrictions:

1. Reduction of the cavity volume due to thermal expansion of the plastic zone with cavity temperature rise.

2. Limitation of extension of the plastic zone due to the earth's surface.

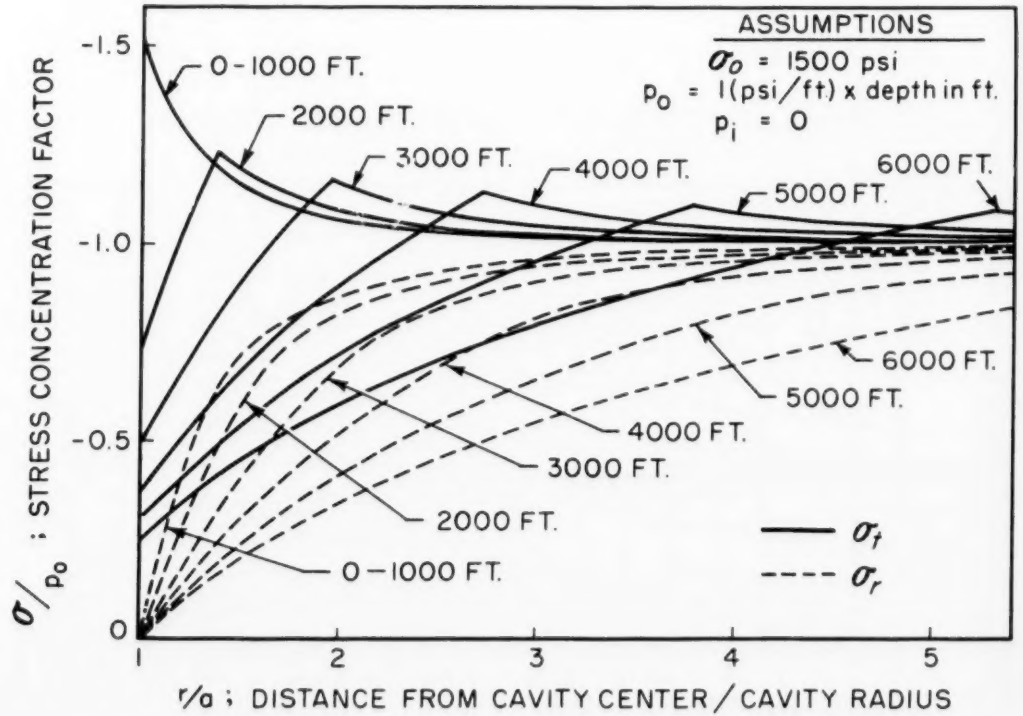

FIG. 20 STRESS DISTRIBUTION IN PLASTIC AND ELASTIC ZONES AROUND SPHERICAL SALT CAVITY AT VARIOUS DEPTHS.

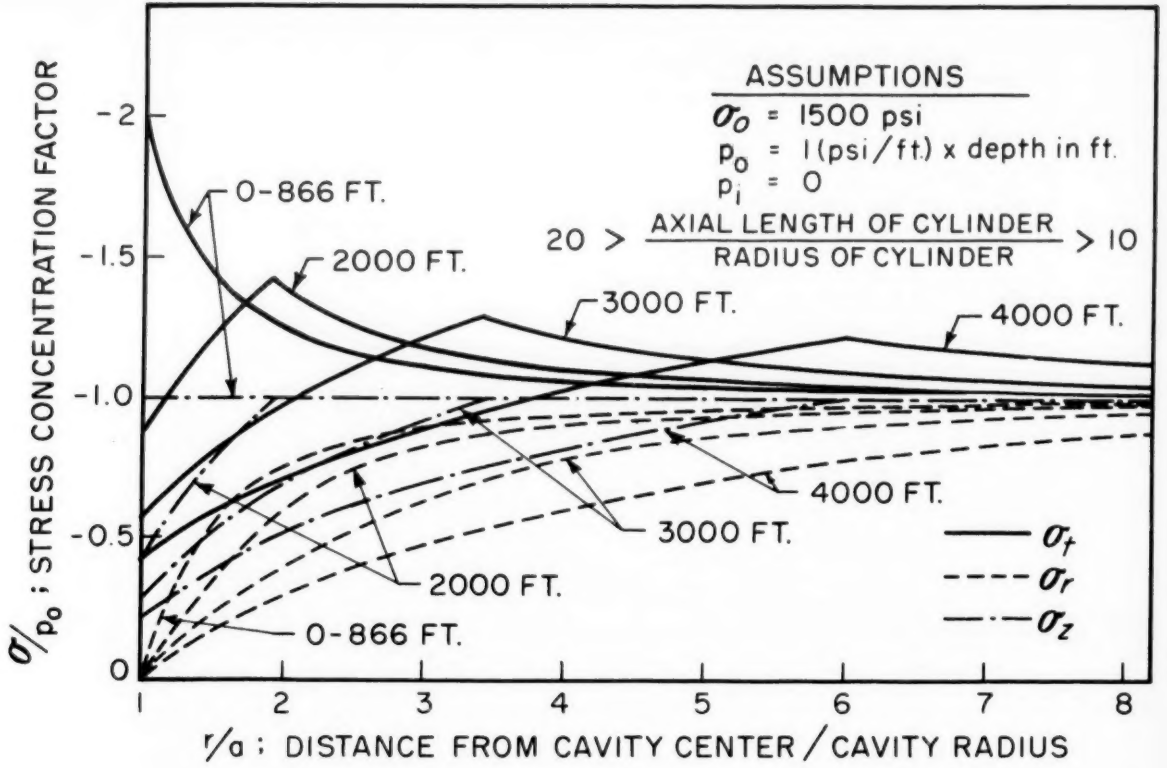

FIG. 21 STRESS DISTRIBUTION IN PLASTIC AND ELASTIC ZONES AROUND CYLINDRICAL SALT CAVITY AT VARIOUS DEPTHS.

3. Reduction of the cavity volume due to the relaxation of salt in the plastic zone.

As analyzed in the following discussion, the reduction of the cavity volume due to the thermal expansion will be an actual limit to the use of the cavity for waste storage.

Some observations in the Grand Saline Salt mine, Texas, seem to agree with the conclusion of this plasticity analysis.

The effect of elevated temperature in the cavity upon the development of a plastic zone and the reduction of cavity space due to the thermal expansion and plastic flow of salt are analyzed in the following chapter.

ZONES AROUND CYLINDRICAL SALT CAVITY AT VARIOUS DEPTHS.

Thermal Stress

Thermal stress is produced whenever a local temperature rise occurs in an underground formation. This thermal stress is produced around a salt cavity when the cavity temperature rises above the surrounding earth temperature. The cavity temperature rise occurs as the result of radioactive decay of the fission products in the stored waste. This thermal stress can be determined by calculating the temperature distribution around the cavity which is a function of the cavity geometry and the cavity temperature rise.

Because of the extremely slow change in the rate of heat emission from the waste, the heat flowing from the cavity to the surrounding environment approaches a state of thermal equilibrium; therefore, the temperature at any point around the cavity at any time can be assumed to be a constant.

Mathematical analysis of thermal stress distribution around a salt cavity with elevated temperature is easily obtainable only for very simple

geometrical configurations such as a sphere or cylinder with a long axial length.

The actual thermal stress distribution around spherical and cylindrical salt cavities due to cavity temperature rise can be calculated by the use of the proper values of the three coefficients of salt; namely, Young's modulus, Poisson's ratio and the coefficient of linear expansion.

In the test data reported herein, the Young's modulus of salt was found to be about 150,000 psi at room temperature and the Poisson's ratio of salt to be about 0.5. The coefficient of linear expansion of salt used in these experiments was $22.4 \times 10^{-6}/F°$ at room temperature.

By using these figures, the temperature and thermal stress distributions around the cavity were computed as shown in Figs. 22, 23, and 24. To use the diagrams, thermal stress can be determined by multiplying the temperature rise in the cavity with the unit thermal stress increase obtained from the diagrams.

In practice, the calculated thermal stress should be superimposed on the structural stress to obtain the overall stress distribution around a salt cavity. Thus, a rational estimate of the stress distribution around a salt cavity can be made from a thermal and structural analysis of spherical and cylindrical cavities.

Development of Structural Equilibrium Equations for Design Principle

In order to design an underground salt cavity for the storage of reactor fuel waste, the relationship among cavity temperature rise, structural loading and cavity volume reduction must be established. By introducing both thermal

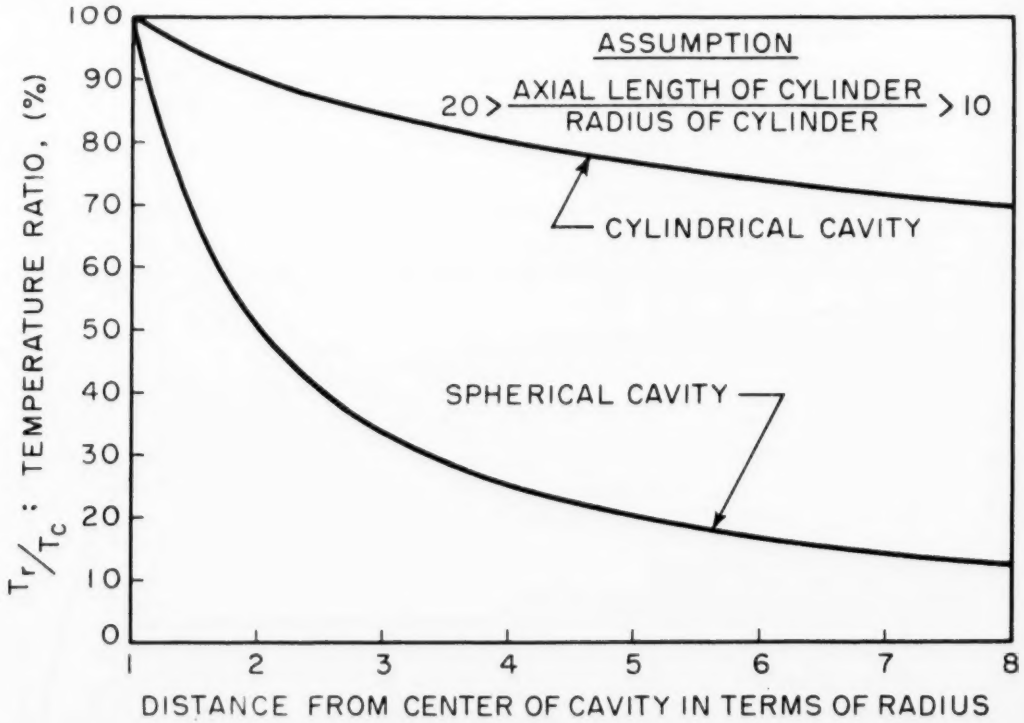

FIG. 22 TEMPERATURE DISTRIBUTION AROUND SPHERICAL AND CYLINDRICAL CAVITIES WITH STEADY STATE HEAT FLOW.

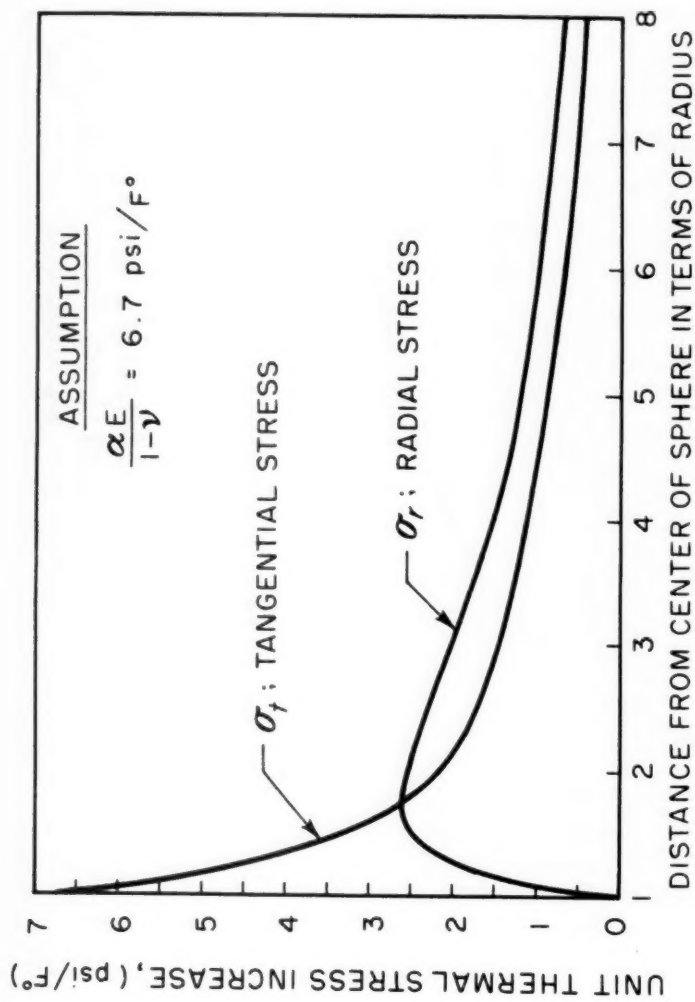

FIG. 23 DISTRIBUTION OF UNIT THERMAL STRESS INCREASE AROUND SPHERICAL SALT CAVITY PER UNIT TEMPERATURE RISE IN CAVITY.

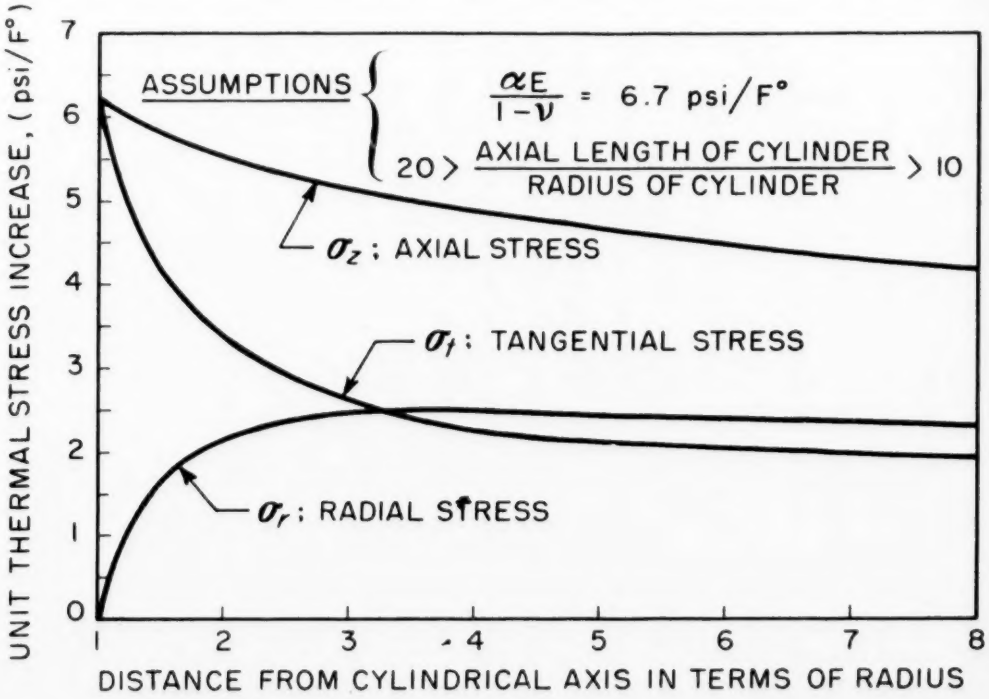

FIG. **24** DISTRIBUTION OF UNIT THERMAL STRESS INCREASE AROUND CYLINDRICAL SALT CAVITY PER UNIT TEMPERATURE RISE IN CAVITY.

stress and structural stress into the plasticity analysis, development of plastic zone around cavities of various temperatures is obtained as given in Fig. 25. As shown in this diagram, the greater the cavity temperature rise and the deeper the cavity depth, the greater the plastic zone. It is to be noted that the plastic zone will be greater in a cylindrical cavity than in a spherical cavity.

The salt in the plastic zone will be subject to thermal expansion followed by plastic flow of the salt. This flow results in reduction of cavity volume. By relating the cavity volume reduction to the development of the plastic zone, the structural equilibrium equations for both spherical and cylindrical cavities are obtained as shown in Figs. 26 and 27. This is the structural equilibrium condition which the cavities will attain as an ultimate stable condition. In these diagrams, structural equilibrium should occur some place between the maximum and the minimum closures, given by solid and dash lines, depending upon the creep velocity of the cavity. The reduction of the cavity volume due to the elevated temperature is one of the most important practical limitations upon the use of the salt cavity. In other words, the cavity may be used successfully for waste disposal if the cavity temperature rise is controlled.

Strength of Salt

The reaction of different materials to a given stress condition varies depending entirely upon their structural properties. The structural properties can usually be described sufficiently with a few fundamental coefficients such as Young's modulus, Poisson's ratio, and yielding stress. These coefficients are essential for any application of the results of the theoretical analysis.

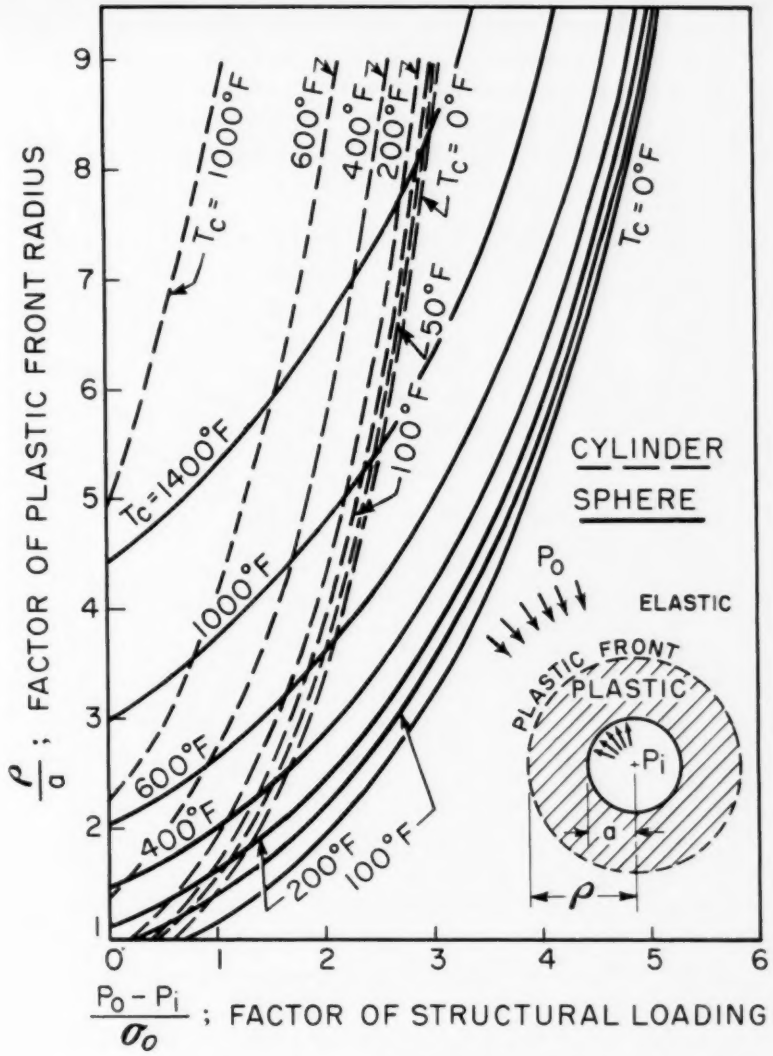

FIG. 25 DEVELOPMENT OF PLASTIC ZONE AROUND SPHERICAL AND CYLINDRICAL CAVITIES WITH VARIOUS TEMPERATURE RISES.

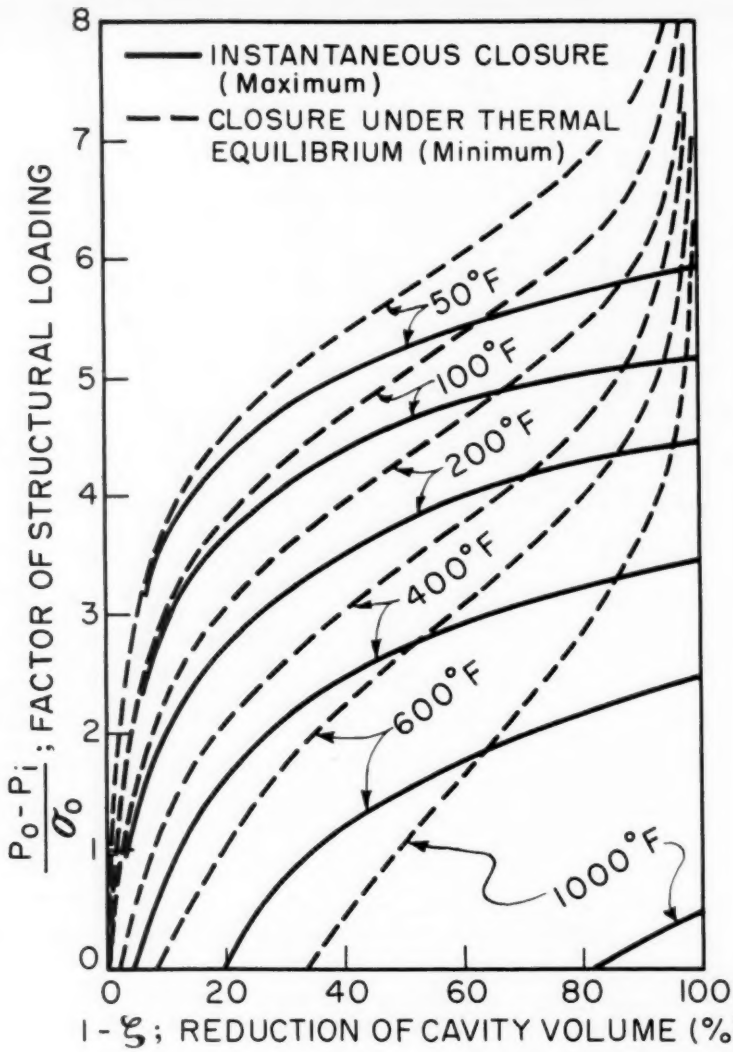

FIG. 26 RELATION BETWEEN ULTIMATE VOLUME REDUCTION AND STRUCTURAL LOADING WITH VARIOUS TEMPERATURE RISES IN SPHERICAL CAVITY.

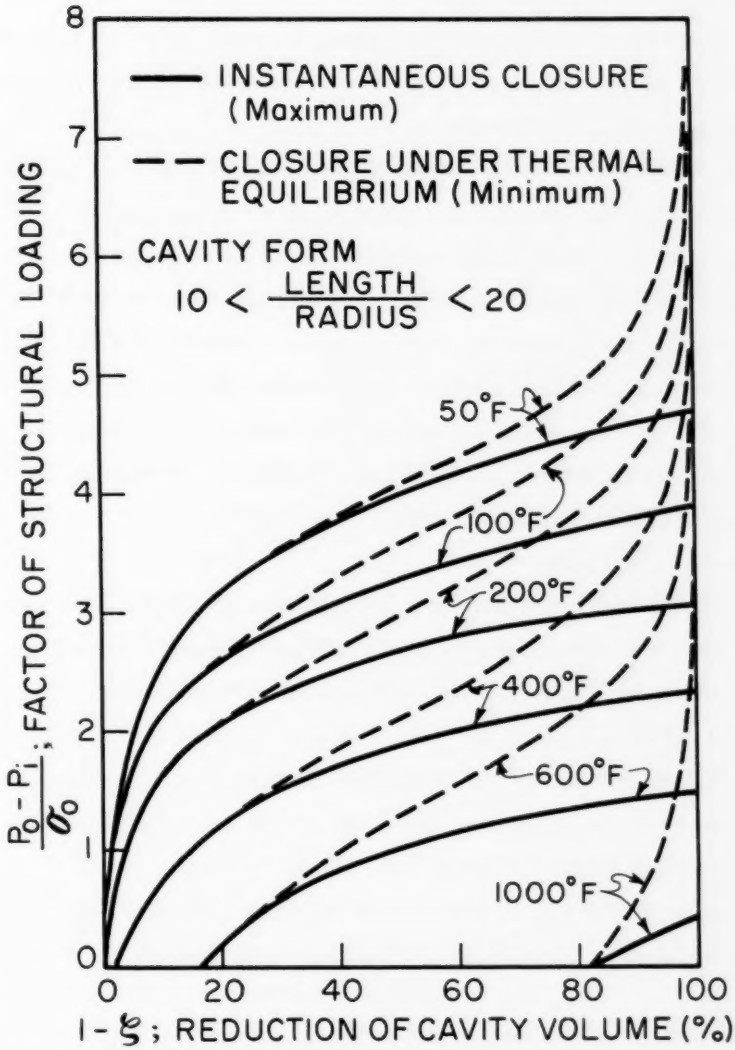

FIG. 27 RELATION BETWEEN ULTIMATE VOLUME REDUCTION AND STRUCTURAL LOADING WITH VARIOUS TEMPERATURE RISES IN CYLINDRICAL CAVITY.

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There are several ways to define the strength of a material. The maximum strength (or ultimate strength) has been the most popular way of describing the strength of a material. The maximum strength of salt was previously determined to be 2,300 psi with a standard deviation of 200 psi by statistical extrapolation.

The determination of mechanical properties of a material is generally affected by the environmental conditions in which the material is found. The strength of salt is influenced by temperature and stress conditions in which the salt occurs. At room temperature, salt is brittle and fractures under uniaxial compression. Salt becomes more ductile, however, and yields plastically under high temperature or triaxial compression. The strength of ductile salt can be analyzed by using the theories of octahedral shearing stress and the Mohr's envelope technique. However, neither can explain every case of brittle failure at ordinary temperature when there is little or no triaxial compression.

The proper development of a "strength theory" of salt under various conditions was sought here because a salt cavity might be exposed to a wide range of temperature and stress conditions from the time of its creation until actual storage of radioactive wastes. The effect of triaxial compression on the strength of salt was studied by constructing Mohr's envelopes from various test data. The important effect of the mean principal stress was investigated by the confinement test and the cylindrical cavity test. All the experimental results were finally evaluated to determine the equivalent plastic limit (σ_o) in relation to the confining pressure.

Summary

A summary of the findings regarding the strength of salt and the salt cavity are:

1. The real cause of failure or creep of a salt cavity is not maximum stress, but mainly the difference between the maximum and minimum stress, acting on the cavity boundary.

2. This stress difference is proportional to the maximum shearing stress, which is the direct cause of failure or creep of the salt cavity.

3. When the maximum shearing stress reaches a critical point on the boundary of the cavity, the salt yields by plastic deformation rather than failing by brittle fracture.

4. Salt can withstand shock loading without suffering structural damage.

5. Mohr's theory of rupture cannot be applied to the strength of the salt under triaxial stress, because the theory does not consider the mean principal stress, which has a great influence upon the strength of salt.

6. The theory of octahedral shearing strength cannot be directly applied to the strength of salt if the salt is not ductile.

7. Both strength theories become applicable whenever temperature surpasses 500° F., or the mean of triaxial stresses exceeds 5,500 psi.

8. If the internal pressure in the cavity can be controlled, a salt cavity can be created and kept stable at any depth.

9. If the internal pressure of the cavity is atmospheric, the maximum depth at which no appreciable plastic flow occurs is determined by the form of the cavity and the σ_o value. This σ_o value is an equivalent of the plastic limit of salt, assuming that salt is ductile.

10. The σ_o value can be experimentally determined by testing the creep closure of an ideal hollow cylinder confined by a steel cylinder.

11. Although no precise experimental determination of the σ_o has been completed, the interpretation of the previous experimental results indicates that the σ_o value should occur between 1,000 and 2,000 psi, at a temperature up to 770° F.

12. At the elevated temperatures used in the uniaxial compression test, the σ_o value can be determined.

Test Devices

Two major test devices were constructed to study the structural properties of salt. The individual factors were first studied; then the comprehensive effects of those factors were correlated with the use of high pressure test devices. High pressure cells were developed so that tri-axial stress conditions could be created. If a standardized specimen is fitted tightly into the cells, the mean principal stress of $1/2 (\sigma_{max} + \sigma_{min})$ can be developed. Also, if the specimen is slightly smaller than the standardized size, the compression and extension effects can be studied. The device is shown in Fig. 28.

In addition to this tri-axial compression capability, the high pressure cells are also useful in studying the effect of solutions and heat on the structural property of salt. A test specimen can be exposed to a solution in the cell by replacing the confining oil with any solution. For studying the effect of elevated temperatures, heat is supplied by a 750 watt heating element attached to the cell. The temperature of the salt in the cell is measured by a thermocouple installed in the bottom plate.

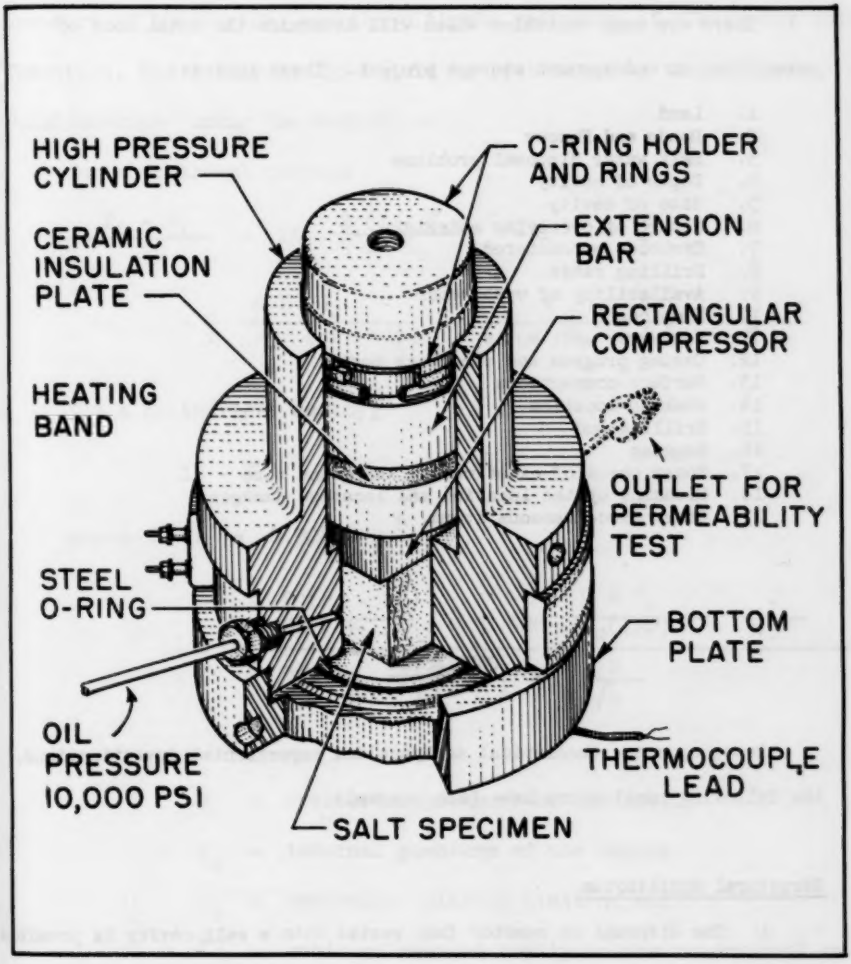

FIG. 28 HIGH PRESSURE CELL ASSEMBLY.

Test devices for measuring actual mine closures consisted of iron plugs imbedded in the salt, aluminum measuring rods, and dial gages.

Economic Factors

There are many variables which will determine the total cost of completing an underground storage project. These include:

1. Land
2. Roads and Fences
3. Salt water disposal problems
4. Depth of cavity
5. Size of cavity
6. Nature of overlying material
7. Troubles encountered
8. Drilling rates
9. Availability of wash water
10. Pump costs
11. Work-over unit costs
12. Casing program and cementing costs
13. Surface connections
14. Staking location
15. Drilling mud
16. Logging
17. Sonar check of exact cavity shape and size
18. Cleaning up the location and location damages
19. Other miscellaneous items.

CONCLUSION

Based upon the theoretical analyses and experimental investigations, the following conclusions have been reached:

Structural Equilibrium

1. The disposal of reactor fuel wastes into a salt cavity is possible because a properly designed cavity establishes an ultimate structural equilibrium.

2. The ultimate strucutral equilibrium can be determined by the relationships found to exist among four major variables: the cavity temperature rise, the overburden pressure of the cavity, the internal pressure of the cavity, and the ultimate reduction of the cavity volume. Therefore, the cavity design should be restricted by the following relationships among the variables:

for a spherical cavity:

$$\frac{P_o - P_i}{\sigma_o} = \ln \frac{2(1-\zeta)}{9\alpha T_c} + 1 + \frac{2}{3} - \frac{2}{3}\frac{\alpha E T_c}{\sigma_o (1-\nu)} \qquad \frac{9 \alpha T_c}{9\alpha T_c + 2(1-\zeta)}$$

for a cylindrical cavity:

$$1 - \zeta = 3 \alpha T_c \ (Ae^B - 1.072)$$

where $A = 0.928 - \frac{B}{13.8}$

$$B = \frac{\frac{2(P_o - P_i)}{\sigma_o} + 0.928 \frac{\alpha E T_c}{\sigma_o(1-\nu)} - \frac{2}{\sqrt{3}}}{0.072 \frac{\alpha E T_c}{\sigma_o (1-\nu)} + \frac{2}{\sqrt{3}}}$$

P_o = overburden pressure of the cavity

P_i = internal pressure of the cavity

σ_o = equivalent plastic limit of salt

ζ = ratio of reduced cavity volume to original one

ν = Poisson's ratio of salt

α = coefficient of linear expansion of salt

T_c = cavity temperature rise above ambient temperature

E = Young's modulus of salt

$$10 < \frac{\text{LENGTH OF CYLINDER}}{\text{RADIUS OF CYLINDER}} < 20$$

3. Ultimate structural equilibrium occurs because a plastic zone may develop around the cavity. This plastic zone may possibly extend to contiguous formations. The radius of the plastic zone, ρ, can be calculated from the following equations:

for a spherical cavity:

$$\frac{P_o - P_i}{\sigma_o} = 2 \ln \frac{\rho}{a} + \frac{2}{3} - \frac{2}{3} \frac{\alpha E T_c}{\sigma_o (1 - \nu)} \quad \frac{a}{\rho}$$

for a cylindrical cavity:

$$\frac{P_o - P_i}{\sigma_o} = \frac{2}{\sqrt{3}} \ln \frac{\rho}{a} + \frac{1}{\sqrt{3}} - \frac{\alpha E T_c}{2 \sigma_o (1-\nu)} \quad (0.928 - \frac{\ln \rho - \ln a}{6.91})$$

where a = radius of the cavity (spherical or cylindrical)

$$10 < \frac{\text{LENGTH OF CYLINDER}}{\text{RADIUS OF CYLINDER}} < 20$$

4. The reduction of cavity volume imposes a practical limitation upon the depth of the cavity and the permissible temperature rise in the cavity. This reduction in volume is mainly caused by thermal expansion rather than structural expansion of the salt in the plastic zone.

5. The ultimate structural equilibrium of an irregularly shaped cavity can be approximated by comparisons with ideal spherical and cylindrical cavities. A cavity of irregular form would probably be as stable as a cavity of ideal form, since minor irregularities will not produce higher stress concentration because of the plastic nature of salt.

6. In general, salt cavity storage becomes impractical when the cavity temperature exceeds 500°F. The temperature rise in the cavity is restricted by two factors: One is the reduction of the cavity volume due to the thermal expansion of salt; the other is the increase of the waste vapor pressure beyond the overburden pressure.

7. The loading condition of a salt cavity is best described by the term "factor of structural loading" which is defined as:

$$X = \frac{P_o - P_i}{\sigma_o}$$

where X = factor of structural loading

P_o = overburden pressure of the cavity

P_i = internal pressure of the cavity

σ_o = equivalent plastic limit of salt

The structural load on the cavity is minimized if the internal pressure is equal to the overburden pressure. The cavity can be fractured if the internal pressure exceeds the overburden pressure.

8. The elastic theory can be applied for stress analysis around a salt cavity when the octahedral shearing stress around the cavity is less than its maximum value for salt.

9. The theory of plasticity must be introduced into the analysis whenever the stress around the cavity exceeds the maximum octahedral shearing stress of salt.

10. Increased temperatures not only create thermal stress but also extend the plastic zone around the cavity.

11. Stress concentration around the cavity is independent of the size of the cavity. However, the extent of the stress concentration away from the boundary is proportional to the size of the opening.

12. If no plastic zone develops, the geometry of a cavity determines the stress concentration around the boundary where critical stress and maximum shearing stress always occur.

13. Generally, the depth of the cavity determines the overburden pressure on the cavity; therefore, the shallower the depth, the less the external load. However, the thickness of the overburden layer above the cavity should be at least 3 times the cavity diameter.

14. The experimental results of creep measurement in the Grand Saline salt mine agree with the theoretical analysis. The empirical creep equation developed for the mine opening indicates that creep in the mine, at the depth of 700 ft., will practically disappear within a decade after the creation of the opening; and that the total amount of the closure resulting from creep will not exceed several percent of the opening diameter or height.

Young's modulus	0.142×10^6 psi
with the standard deviation	0.03×10^6 psi
Poisson's ratio	
with the compressive stress up to 300 psi	0.25 - 0.5
with the compressive stress over 300 psi	0.5

The wide variation in the previously reported figures regarding the fundamental properties of salt is probably due to the varied testing procedures used by previous investigators, besides some difference in the properties of the tested salt.

2. The stress condition in a salt formation becomes hydrostatic at a depth approximately below 300 ft.

3. Salt specimens described herein are structurally isotropic and homogeneous regardless of their previous experience in stress and strain.

4. The total creep of salt consists of instantaneous, transient, and steady state creeps and can be formulated with respect to time:

$$\epsilon_o = \alpha + \beta \log t + \gamma t$$

where ϵ_o = total creep

α = instantaneous creep

β = the coefficient of transient creep

γ = the coefficient of steady state creep

5. Heat has the most significant effect upon the structural property of salt. The creep rate of salt is increased 75 times with temperature increase of 80° F to 770 ° F. This means that a salt cavity reaches its ultimate structural equilibrium much faster if the cavity temperature is elevated. The equivalent plastic limit of salt is reduced to 1,000 psi with temperature increase of 80° F to 500° F; thereafter, the equivalent plastic limit remains nearly constant at around 1,000 psi while the temperature approaches the melting point of salt. Heat affects Young's modulus and the coefficient of linear expansion of salt very slightly.

6. Gamma irradiation of salt up to 20×10^6 rep has no effect upon the fundamental structural properties of salt. However, the irradiated salt exhibits a slight reduction in the rate of transient creep.

7. The maximum strength of salt is not affected by its submersion for periods of one month in light mineral oil, reactor fuel waste substitute (nitrate waste), and saturated salt water.

8. Salt submerged in saturated water exhibited some increase in creep although no similar effect was found in salt submerged in the reactor fuel waste substitute.

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Representative HOLIFIELD. We will ask Mr. R. E. Tomlinson, of Hanford, to come to the table.

Mr. Tomlinson is from Hanford working for General Electric and will also give us some testimony on high-level waste disposal.

STATEMENT OF R. E. TOMLINSON,[7] MANAGER, ADVANCE PROCESS DEVELOPMENT CHEMICAL PROCESSING DEPARTMENT

Mr. TOMLINSON. Thank you, Mr. Chairman.

My name is R. E. Tomlinson; I am employed by the General Electric Co. at the Hanford atomic products operation in Richland, Wash., as manager of advance process development in the chemical processing department.

I have been asked to comment on the removal of specific radioisotopes from the chemical wastes of the reactor fuels reprocessing operations as such removal might affect the disposal of these wastes.

The aqueous chemical wastes from the reprocessing of irradiated fuels are composed primarily of nonradioactive materials. In the high-level wastes produced in the past, for example, each pound of fission products has been associated with about 1,000 to 10,000 pounds of inactive salts, excluding the water which is used to facilitate the movement of the wastes and to aid in the dissipation of the heat of radioactive decay. If the radioisotopes requiring containment could be quantitatively removed for separate containment, the remaining materials might be treated as normal industrial wastes.

It would appear, then, that a cheap method of isolating the isotopes of concern would permit cheaper waste disposal methods, improved certainty of containment, or both. This possibility has been under study at many AEC sites for several years.

Let us consider the specific fission products of concern. Table 1, included as an attachment to this testimony, presents the approximate concentrations and half lives of the significant fission products to be found in the chemical waste from the reprocessing of a typical irradiated fuel. The assumed conditions of irradiation and reprocessing

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Technical adviser, U.S. delegation, Peaceful Uses of Atomic Energy, Geneva, 1955.
Work history:
1939 to 1941: Laboratory assistant, operator, dye works, E. I. du Pont Co., Deepwater, N.J.
1941: Supervisor trainee, TNT, E. I. du Pont Co., Millington, Tenn.
1941 to 1944: Foreman, Kankakee ordnance works, E. I. du Pont Co., Joliet, Ill.
1944: Supervisor trainee, Clinton Laboratories, E. I. du Pont Co., Oak Ridge, Tenn.
1944 to 1945: Construction checking, Hanford works, E. I. du Pont Co., Richland, Wash.
1945: Shift supervisor, TNT, Oklahoma ordnance works, E. I. du Pont Co., Pryor, Okla.
1945 to 1946: Foreman, Azo Analytical Laboratory, Chambers works, E. I. du Pont Co., Deepwater, N.J.
1946 to 1948: Chemical engineer, technical section, General Electric Co., Richland, Wash.
1948 to 1951: Chemical engineer, design and construction section, General Electric Co., Richland, Wash.
1951 to 1953: Supervisor, production planning, technical section, General Electric Co., Richland, Wash.
1953 to 1954: Head, hot semiworks, technical section, General Electric Co., Richland, Wash.
1954 to 1956: Head, process planning, technical section, General Electric Co., Richland, Wash.
1956 to present: Manager, advance process development, chemical processing department (currently acting manager, production, chemical processing department), General Electric Co., Richland, Wash.
Professional societies: Member, American Institute of Chemical Engineering.

are significant only to the extent that they represent possible conditions. Of the 15 nuclides listed in table 1, all but 6 decay to innocuous levels within the first 6 years. These six are listed on table 2 which has been prepared in the form of a chart. During the 6 years' storage period, the rate of heat generation from radioactive decay also decreases about tenfold from the rate associated with the irradiated fuel after, say, 90 days cooling. The process and engineering problems involved in waste treatment operations are thus greatly simplified by merely waiting a few years. It is, therefore, probable that the high-level wastes will be stored for a few years prior to any efforts to process them for improved certainty of long-term containment. More than a thousand years would be required, however, for all of the fission products to decay to levels considered safe for drinking purposes.

TABLE 1.—*Comparative amount of fission products in waste from the reprocessing of irradiated fuels*

Nuclide	Concentration [1] /μc/cc	T ½ years	MPC [2] /μc/cc of H_2O	Years decay to reach MPC	Ratio of concentration to MPC after 6 years decay
Sr^{89}	1.6×10^4	0.148	7×10^{-3}	4.1	
Sr^{90}	2.3×10^4	28	8×10^{-7}	1,000	2×10^{10}
Y^{91}	2.3×10^5	0.159	3×10^{-4}	4.7	
Zr^{95}	2.7×10^5	0.178	6×10^{-4}	5.1	
Ru^{103}	8.5×10^4	0.110	9×10^{-4}	2.9	
Ru^{106}	7.2×10^4	1.0	1×10^{-4}	30	1×10^7
Te^{129}	3.1×10^3	0.090	2×10^{-4}	2.2	
Cs^{137}	2×10^4	30	2×10^{-3}	700	8×10^6
Ba^{140}	5.2×10^3	0.035	3×10^{-4}	0.8	
Ce^{141}	9×10^4	0.088	4×10^{-1}	1.6	
Pr^{143}	5.5×10^3	0.038	5×10^{-4}	0.9	
Ce^{144}	6.9×10^5	0.780	1×10^{-4}	26	3×10^7
Nd^{147}	1.6×10^3	0.032	6×10^{-4}	0.7	
Pm^{147}	9×10^4	2.6	2×10^{-3}	66	9×10^6
Sm^{151}	5.6×10^2	80	8×10^{-3}	1,300	7×10^4

[1] Assumes the fission products from 1 ton of irradiated (2,500 MWD/T at 5 MW/T) uranium (1.2 percent U^{235} before irradiation) are segregated in 100 gallons of waste 90 days after reactor discharge.

[2] The maximum permissible concentration (MPC) for each nuclide in potable water is given for each parent in equilibrium with its radioactive daughters. These values were taken from HW-25457, Rev. 1, Manual of Radiation Protection Standards, Hanford Atomic Products Operation, May 1, 1957.

TABLE 2.—*Long-lived fission products in high level wastes*

Isotope	Years required to decay to MPC for drinking water	Number of times above MPC 6 years' storage
Strontium 90	1,000	20,000,000,000
Ruthenium 106	30	10,000,000
Promethium 147	66	9,000,000
Cesium 137	700	8,000,000
Cerium 144	26	3,000,000
Samarium 151	1,300	70,000

(Other fission product isotopes in this hypothetical waste would decay to below the MPC in less than 6 years.)

Five of the six isotopes requiring long-term containment have potential value to industry. Uses are being actively developed for strontium 90, ruthenium 106, cesium 137, cerium 144, and promethium 147, and processes are being developed for their recovery. A pilot plant is now in operation at Oak Ridge recovering some of these iso-

topes. This aspect of the picture will be discussed by other witnesses later.

The recovery of specific isotopes for beneficial use is a quite different problem from that of removing them to facilitate waste disposal. For a recovery process, a yield of 90 percent might be quite satisfactory, since the primary goal would be the isolation of selected isotopes at low cost. If the goal were to permit the residue to be treated as a conventional industrial waste, however, essentially complete removal would be required for all of the elements of concern. For these purposes, any plutonium or other transuranic elements present must also be removed with comparable efficiency. Microscopic "losses" ranging as low as 1 part per 20 billion would have to be simultaneously achieved for them any elements of widely different chemical characteristics. Achieving this simultaneous high recovery of such chemically diverse products is without precedent in the chemical industry.

Several processes are being developed which are theoretically capable of obtaining the indicated removal of individual isotopes. For example, cesium can be adsorbed on phenolic resins (7), or it can be extracted into organic solvents by association with extractable heavy metal complexes (8); strontium can be removed by solvent extraction using di-2-ethyl hexyl phosphoric acid as the solvent (9); plutonium can be removed by solvent extraction using tributyl phosphate or tri-n-lauryl amine (10) as the solvent, or it can be removed by ion exchange (11); any of the elements can be removed by foam separation after addition of selected surface active agents (12). Much additional development would be required before any of these processes could be relied upon to give the required yields on a production basis.

Let us assume that processes can be developed for the near quantitative removal of all of the ions of concern. The combination of required processing steps would almost certainly involve many sequential operations requiring close operational control in an extremely high radiation field. The complexity of the process and engineering problems are such that the waste treatment facilities would probably be as expensive to construct and operate as the fuel reprocessing plant itself.

For those fuels which contain little or no nonfissionable materials (such as aluminum or zirconium), the reprocessing can be modified such that most of the advantages of isotope removal from the waste can be realized by simple means. This approach consists of tailoring the separations processes to minimize the quantities of inactive salts in the chemical wastes. For example, we are learning how to recover fissionable fuel materials without dissolving the cladding materials, to minimize the use of process reagents by recycling waste streams within the process, to use process reagents that can be subsequently removed as gases, and to minimize the corrosion of the process equipment. We hope by these means to decrease the quantity of inactive salts in the chemical wastes such that each pound of fission product is associated with perhaps 20 pounds of inactive salts. After a few years of interim storage, the total wastes might then be calcined to a glassy solid for increased certainty of long-term containment at reasonable cost while avoiding the intrinsically expensive

operation of removing radioactive materials in a near quantitative manner.

In my opinion, efforts to remove specific isotopes from the chemical wastes now being produced should therefore be directed toward, and justified by, the recovery of the isotopes for beneficial use. Such recovery would reduce the problem of heat dissipation in the residual wastes, but would have little effect on the overall cost of waste containment.

(References submitted by Mr. Tomlinson follow:)

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Representative HOLIFIELD. Are there any questions?

Chairman DURHAM. From your statement, Doctor, it is very evident that it will take a thousand years to clear up a stream in case it got contaminated. It is very important that we do not get it mixed up with our water supply to the point where that will become a tremendous problem.

Mr. TOMLINSON. That is correct.

Chairman DURHAM. A person might not want to wait a thousand years to get a drink of water.

Representative HOLIFIELD. Thank you very much, Mr. Tomlinson.

(Additional papers submitted for the record follow:)

REMOVAL OF FISSION PRODUCTS FROM HIGH LEVEL RADIOACTIVE WASTE SOLUTIONS

by

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Work Area: Problem Leader for Waste Treatment and the separation of elements by Ion Exchange

ABSTRACT

Ion exchange, electrolysis, solvent extraction, scavenging and precipitation have been studied as methods for removing radioactive isotopes from high level wastes. Process development has been limited to laboratory research and small scale engineering tests so that the efficiency of these methods in large scale operations is unknown. Emphasis has been placed on removal of strontium^{90} and cesium^{137} which are the most biologically hazardous of the long lived radioisotopes.

Continuous processing of the supernate from neutralized, sodium nitrate or aluminum bearing wastes by ion exchange appears particularly promising. Cesium and strontium have been removed by factors greater than 10^5 and 10^3, respectively. Since about 99% of the strontium remains in the neutralized waste sludge, the over-all removal for strontium was greater than 10^5. These demonstrated purifications would reduce present Oak Ridge National Laboratory wastes to a factor of 100 above drinking water tolerance for strontium and a factor of 10 lower than tolerance for cesium. In these tests, the wastes were reduced to background levels of radioactivity and consequently, higher decontamination factors are expected when higher levels of waste are processed. Similar results are expected to be achieved for the other limiting long lived isotopes, plutonium^{239,240}, curium^{242}, and americium^{241} based on their known solubilities in alkaline solutions and ion exchange characteristics. Removal factors of greater than 10^4 for strontium and cesium are predicted in the treatment of acidic aluminum wastes by the Diban-Ion Exchange process following a scavenging-precipitation step. This process is considered less efficient than the neutralized treatment method since it presents more difficult engineering problems.

Ruthenium^{106} is not removed by ion exchange but can be removed by electrodeposition by factors greater than 200 which is sufficient to reduce the ruthenium concentration to below drinking water tolerance for neutralized high level wastes. Nitrate is eliminated by decomposition in the same cell.

Research studies on a number of organophosphorus acids has shown that di-2-ethylhexylphosphoric acid (D2EHP) may be particularly effective for the selective extraction of strontium from neutralized wastes. A strontium distribution coefficient, $E^{o}_{a}(Sr)$, of 200 and a separation factor of strontium from sodium of 2000 was determined between 0.1 M Na(D2EHP) in Amsco modified with 5% tributyl phosphate and 0.5 M sodium nitrate solution at pH 5. Tributyl phosphate is of only limited interest since it can be used to extract the rare earths, ruthenium, zirconium, and niobium but does not extract cesium or strontium.

INTRODUCTION

Removal of the long lived fission products from high level radioactive wastes will permit disposal of the remaining highly salted waste by conventional industrial methods. In many cases the decontaminated wastes can be disposed directly to the ground, streams or to the ocean without further treatment. The recovered isotopes will be safely isolated in small stainless steel containers for storage or for use by industry as radiation sources. Since many fission products have short half-lives, it is practical to allow them to disappear by decay before processing is undertaken. Calculations[1] have shown that after 5 years of decay, the most hazardous isotopes remaining are strontium^{90}, cesium^{137}, americium^{241}, plutonium^{239,240}, and curium^{242}. In water, strontium^{90} and cesium^{137} are about 3000 times and 10 times more hazardous, respectively, than the third most hazardous isotope, americium^{241}. Decontamination factors of 10^7 and 10^9 (99.9999999% removal) are required to reduce the strontium^{90} content of Oak Ridge National Laboratory (ORNL) intermediate and high level wastes to the maximum permissible concentration (MPC_w) permitted for drinking water.

Some of the treatment methods under development are designed to remove all of the hazardous long lived isotopes by a single technique to achieve a cheap bulk treatment process, whereas other methods selectively remove a single isotope. But in general, development work has concentrated on methods for the removal of cesium^{137} and strontium^{90} because of their more hazardous nature. The mixture of isotopes produced from the bulk treatment methods could be used for irradiation work, stored, or processed further to produce pure isotopes for specific uses. Methods studied at ORNL include ion exchange, precipitation, scavenging, and solvent extraction. In most cases, these methods are in the development stage and have not been evaluated in pilot plants on actual waste streams involving large amounts of radioactivity. Consequently, their true efficiency is unknown. The single exception is the Multicurie Fission Product Pilot Plant[2,3] which produces isotopes from highly radioactive waste streams. The principal processing methods used in this plant are scavenging and crystallization although all of the other methods are employed to some extent for specific applications.

CONCEPT OF A MULTIPURPOSE WASTE PROCESSING FACILITY

In presenting methods for isotope recovery and waste treatment, it is important to empnasize the source of the wastes since in many cases their previous history governs the choice of a treatment method. Figures 1a and 1b show the possible routes that wastes may take considering both actual practice and proposed treatment methods. The heavy lines indicate the preferred paths, based on presently available information and on work under development.

The first step is evaporation to recover the excess acid present in the wastes from the nuclear fuel reprocessing plants. The wastes from the Hanford, Savannah River and Oak Ridge National Laboratory installations, which cummulatively represent over 90% by volume of the wastes, are then neutralized with excess caustic and stored in mild steel or concrete tanks. The single exception is the Idaho Chemical Processing Plant which stores acid wastes in stainless steel tanks. On neutralization, iron and other hydrous oxides precipitate and carry down the colloidal fission products ruthenium, zirconium and niobium. In addition, most of the other fission products such as the rare earths, strontium, zirconium, niobium, ruthenium, and plutonium are insoluble in alkaline, carbonate and sulfate-bearing solutions and are precipitated in the sludge. Most of the non-radioactive bulk chemicals remain dissolved in the alkaline supernatant solution as sodium salts of nitrate, aluminate, sulfate, fluoride and hydroxide. The supernatant solutions also contain approximately 100% of the cesium, 10% of the ruthenium, and 0.1-1% of the strontium, rare earths and transuranics although these percentages can probably be reduced by adding precipitating or scavenging agents such as alum, ferrocyanide, sulfide or phosphate in connection with the neutralization step. This is the extent of large scale waste processing at the present time.

The decontamination factors required to reduce present neutralized ORNL waste supernates to drinking water tolerance are shown in Table 1. It may be seen that factors of 10^5, 10^4, and 5.7 are required for strontium, cesium and ruthenium, respectively. Decontamination factors about 100 times

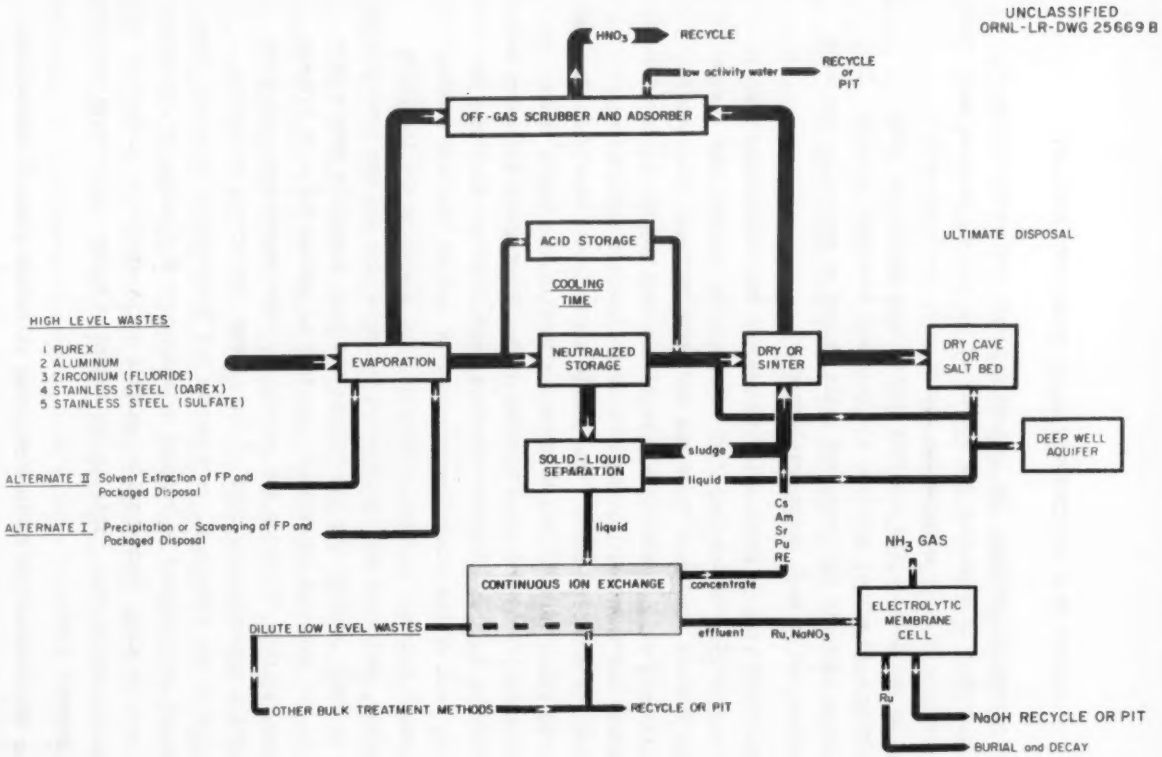

Fig. 1a. Waste Treatment System-Bulk Disposal of Fission Products and Solids. Heavy Lines Indicate the Preferred Paths.

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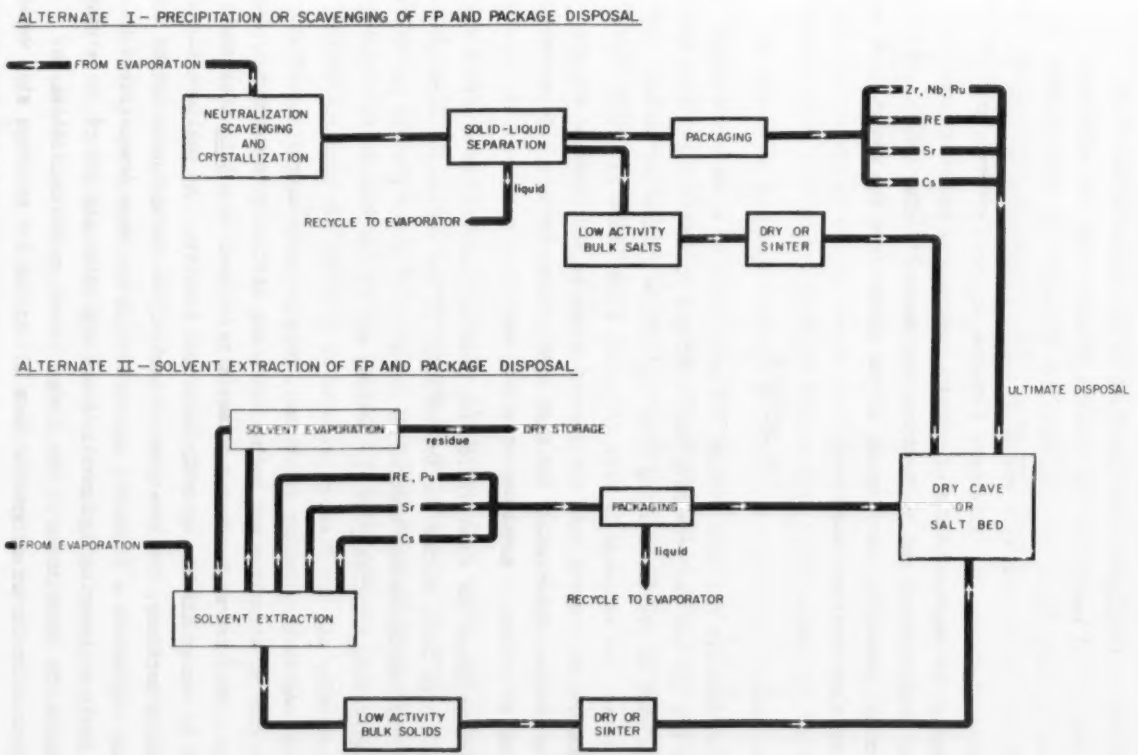

Fig. 1b. Alternate Methods of Fission Product Separation and Package Disposal.

higher will be required for power reactor wastes. Decontamination methods under development are described in the following sections. They propose, 1) removal of the fission products from the supernate from neutralized wastes by continuous ion exchange and electrolysis (Figure 1a), or 2) from acid wastes by solvent extraction as shown by alternate II-Figure 1b. Alternate I-Figure 1b, precipitation and scavenging, is representative of methods employed in the Multicurie Fission Product Plant and is described elsewhere.[2,3] The removal, processing, packaging and disposal of the sludge from the neutralized waste is also described elsewhere.[4]

ION EXCHANGE

Ion exchange has been studied for many years as a waste treatment method but its use has generally been confined to treating low salt bearing streams such as reactor cooling water and dilute laboratory wastes. In these cases, ion exchange is very efficient since there is little inert salt present to compete with the fission products for sites on the ion exchange resin. Conversely, the high level wastes contain high concentrations of sodium, aluminum, or nitric acid.

Ion exchange has shown increasing promise, however, as a method for treating high level wastes for two reasons; 1) the successful development of a truly countercurrent continuous contactor[5,6] which greatly increases the engineering efficiency of the method; and 2) the remarkable affinity of essentially all the fission products of interest for phenolic cation exchange resins in alkaline solutions. This is particularly significant in the case of strontium and cesium which are difficult to extract by other methods. Thus ion exchange can potentially be used as a bulk treatment method to remove all biologically hazardous elements. As outlined in the succeeding sections, the treatment of neutralized waste supernates by ion exchange represents a promising approach which has been demonstrated in small scale engineering equipment. Since approximately 99% of the strontium remains in the waste tanks in the sludge formed on neutralization, a decontamination factor of greater than 10^3 in the ion exchange step would produce an over-all decontamination factor of greater than 10^5 for strontium.

These decontamination factors would reduce the strontium content of the solution to approximately drinking water tolerance in many applications and to 100 times this tolerance for present ORNL wastes. Decontamination factors greater than 10^5 have been demonstrated for cesium which would reduce the activity of present ORNL wastes to a factor of 10 lower than drinking water tolerance.

Of the major radioactivities, only the relatively short lived ruthenium, half life 1.0 yr, is not removed (see Electrolysis in next section). Further large scale tests are required to define the ultimate efficiency of the method for removal of all the major biological hazardous isotopes. Larger decontamination factors are expected when higher level wastes are tested.

The Diban-Ion Exchange method for treating acidic aluminum-bearing wastes, is less highly developed so that its efficiency is difficult to estimate. Based on the limited data available, the process shows some promise for treating aluminum wastes but little promise for treating wastes with high sodium concentrations.

Alkaline Wastes. Ion exchange removal of long lived isotopes from alkaline wastes is attractive in that phenolic cation exchange resins have high distribution coefficients (i.e., one volume of resin can treat many volumes of solution) for cesium, strontium, rare earths and transuranics in such solutions, and the isotopes can be removed from the resin easily with acid. Thus such a treatment is a potential means of decontaminating the bulk alkaline chemical wastes (except for ruthenium) so that they can be discharged to the environment.

Miller and Kline[7] found that cesium is selectively sorbed by phenolic cation exchange groups in the presence of high concentrations of sodium salts in alkaline solutions. Watson and Evans[8] and Heal and Crouch[9] developed a method for cesium recovery from alkaline solutions containing two to four molar sodium using Duolite C-3, a commercially available resin containing both phenolic and methylene-sulfonic groups. Barney[10] also found a high degree of cesium removal from alkaline waste solutions by Duolite C-3, i.e., volumetric distribution coefficients for tracer levels of cesium greater than 100 in a sodium nitrate-aluminate-hydroxide solution containing 4.2 M sodium. Goodall[11] found that phenolic methylene-sulfonic resins

removed cesium from synthetic aluminum bearing Hanford Redox neutralized wastes with a distribution coefficient greater than 50.

Higgins and Messing[12] found cesium distribution coefficients of greater than 100 on Duolite C-3 from a variety of synthetic alkaline waste solutions for trace levels of cesium, with the distribution coefficient dropping to about 50 for cesium concentrations of one gram per liter. Sodium distribution coefficients were approximately unity, so that the cesium-sodium separation factors were approximately equal to the distribution coefficient, i.e., 50 to greater than 100. Trace amounts of strontium and rare earths had distribution coefficients as high or higher than cesium. Distribution coefficients for transuranics were not measured but, judging from their chemistry, should behave like the rare earths. In 3 M HCl regenerant solution, the distribution coefficients are approximately 0.8 for sodium, 2 for strontium and rare earths, and 5.5 for cesium. Thus the resin is readily regenerated and further separation of cesium from strontium and rare earths and of all of these isotopes from sodium is obtained. Sulfuric acid can also be used for regeneration, but not nitric acid since it reacts with the resin.

Higgins and Messing further demonstrated, on a small engineering scale, a continuous ion exchange process for removing the cesium, strontium, and other fission products from alkaline wastes and recovering the cesium for use as a radiation source. They presented a flowsheet, conceptual plant design, and approximate cost estimate for doing this on a large scale using 22,000 curies per day from waste containing one curie of cesium per gallon as a design basis. Using this work as a basis, a flowsheet has been proposed for processing ORNL wastes (Fig. 2).

Higgins and Messing demonstrated waste decontamination factors of greater than 10^4 for cesium and greater than 10^3 for strontium and rare earths. These wastes were reduced to background counting levels, hence the true decontamination factors may have been much higher. From what is known about distribution coefficients and theoretical stage heights there is no reason to believe that any desired decontamination factor cannot be achieved. The cesium to sodium ratio in the cesium product was approximately

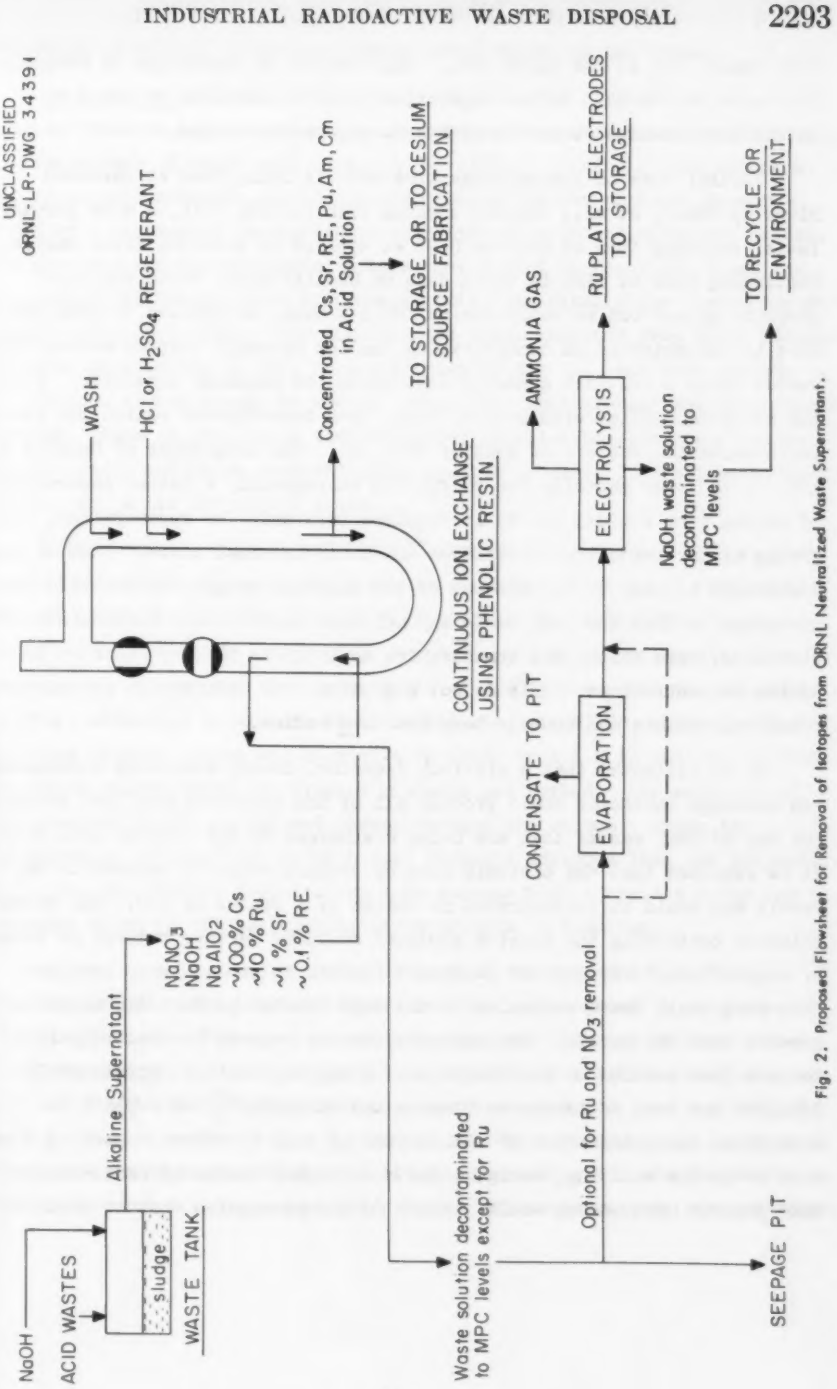

Fig. 2. Proposed Flowsheet for Removal of Isotopes from ORNL Neutralized Waste Supernatant.

2000 times that in the waste feed. This degree of separation is adequate for waste treatment. Better separation could be achieved by using a larger ion exchange contactor with more separation stages.

Further work on ion exchange treatment is being done at Savannah River by Tober, et al., who are working with Duolite C-10, a more porous, faster reacting form of Duolite C-3, as applied to Savannah River wastes. Continuing work at ORNL is being done on Duolite S-30, which contains phenolic groups but no methylene-sulfonic groups, as applied to ORNL wastes. Work is concentrated on demonstrating the ion exchange step on actual ORNL wastes using a one-inch diameter continuous ion exchange contactor. Batch ion exchange studies with Duolite S-30, have demonstrated cesium and barium decontamination factors of greater than 10^5. The advantages of Duolite S-30 are ; 1) a lower capacity for sodium and consequently a better separation of cesium from sodium; and 2) it requires less acid for regeneration, thus giving higher concentration factors and lower chemical costs. Both of these advantages are due to the absence of the sulfonic groups. A possible disadvantage is that the S-30 resin may not have distribution coefficients for strontium, rare earths and transuranics as large as the C-3 resin at high sodium concentrations. This is not a problem with ORNL wastes as they are relatively dilute and contain less than 1 M sodium.

It is estimated that a six-inch diameter, twenty-foot high continuous ion exchange contactor could process all of the approximately 7000 gallons per day of ORNL wastes that are being discharged to the seepage pits (Fig. 2). It is expected that the cationic fission product would be reduced to MPC levels and would be concentrated in volume by a factor of 100. The product solution containing the fission products could be further reduced in volume by evaporation to dryness or processed further to produce pure isotopes. This step would leave ruthenium as the only fission product discharged at greater than MPC levels. The ruthenium can be removed by electrolysis if desired (see section on Electrolysis). A capital cost of approximately $750,000 has been estimated by Blomeke and Stockdale[13] for a plant to accomplish decontamination of ORNL wastes by this flowsheet. Most of this cost is in the building, designed for much higher levels of radioactivity than present ORNL wastes would justify on the assumption that it would be

desirable to be able to process higher level wastes in the future.

Acid Wastes. In early work Blanco, Higgins and Kibbey[14] showed that fission products could be successfully removed from acidic wastes containing a large amount of inert salt, i.e., 1.0 M $Al(NO_3)_3$, using cation exchange. The colloidal ruthenium, zirconium and niobium fission products were removed by a scavenging precipitation with hydrous iron and manganese oxides. The aluminum and fission products were then sorbed on Dowex 50 resin, the aluminum selectively eluted with oxalic acid, and finally the fission products eluted with nitric acid. The aluminum was decontaminated from gross fission products by a factor of 10^3, from plutonium by 5×10^5, and from cesium, strontium and rare earths by 10^3-10^4 (Fig. 3). This process was uneconomical, however, since it involved a stoichiometric relationship between aluminum and the resin and hence required large amounts of resin in addition to large amounts of oxalic and nitric acids.

More recently, Higgins and Wymer[15] proposed an alternate scheme, the Diban-Ion Exchange process, for removing strontium, cesium and rare earths from aluminum wastes (Fig. 4). In this process, the colloidal fission products are removed by scavenging, as above, and the aluminum solution subsequently evaporated to a temperature of 160°C to convert the aluminum nitrate, $Al(NO_3)_3$, to dibasic aluminum nitrate, $Al(OH)_2NO_3$ (Diban). The solution is then cooled and diluted with water to 1.6 M Al and passed into the resin column where the fission products are sorbed. The majority of the aluminum is not sorbed and passes through the column. Since the monopositive $Al(OH)_2^+$ ion is much less strongly adsorbed than the tripositive Al^{+3} ion, the fission products can more successfully compete for the ion exchange sites in the presence of large amounts of aluminum.

An oxalic acid scrub solution is used to decrease the amount of aluminum sorbed. The aluminum waste can then be discarded or evaporated to 6 M $Al(OH)_2NO_3$ if further storage is required. Storage in this case would require 300-400% less volume than the present practice of storing at 1.5 to 2.0 M $Al(NO_3)_3$. The fission products are removed from the resin by an 3 M ammonium carbonate-0.1 M ammonium citrate solution. The ammonium carbonate can be recovered for recycle by steam stripping and the remaining fission product solution evaporated for low volume storage or the individual

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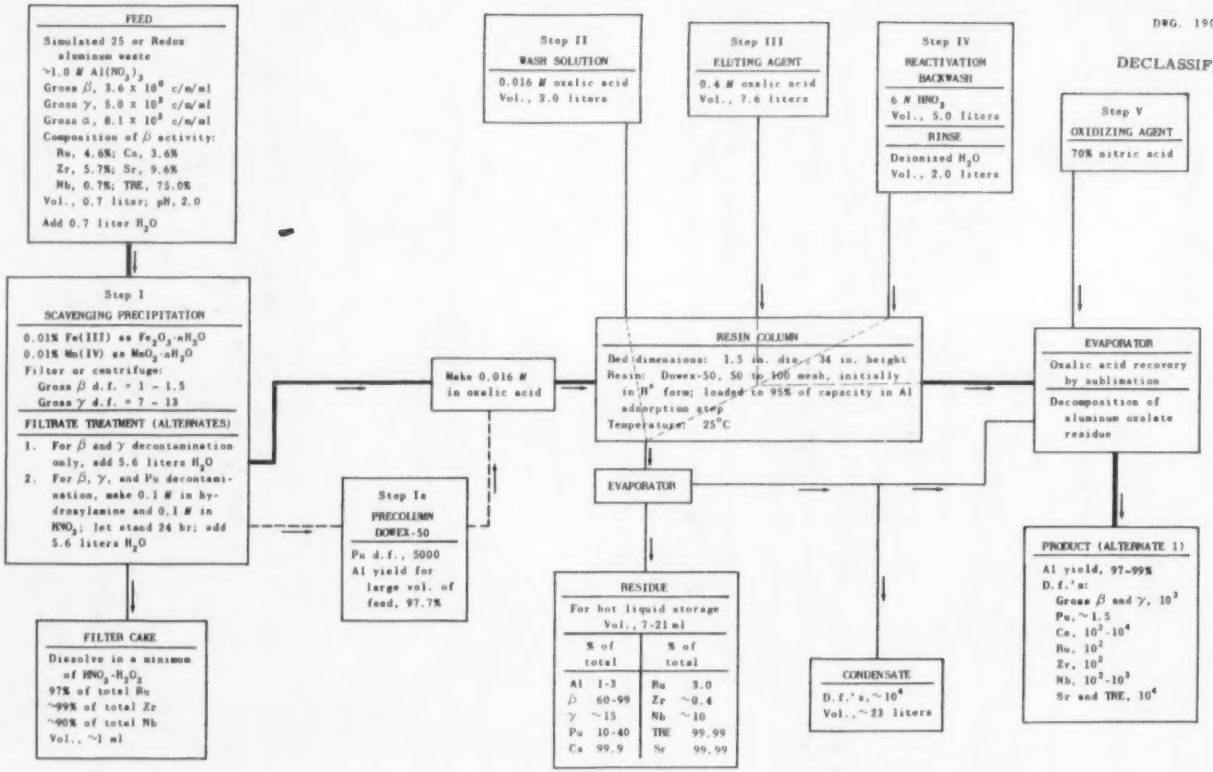

Fig. 3. Removal of Isotopes from Acidic Aluminum Bearing Wastes by Ion Exchange.

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Fig. 3. Removal of Isotopes from Acidic Aluminum Bearing Wastes by Ion Exchange.

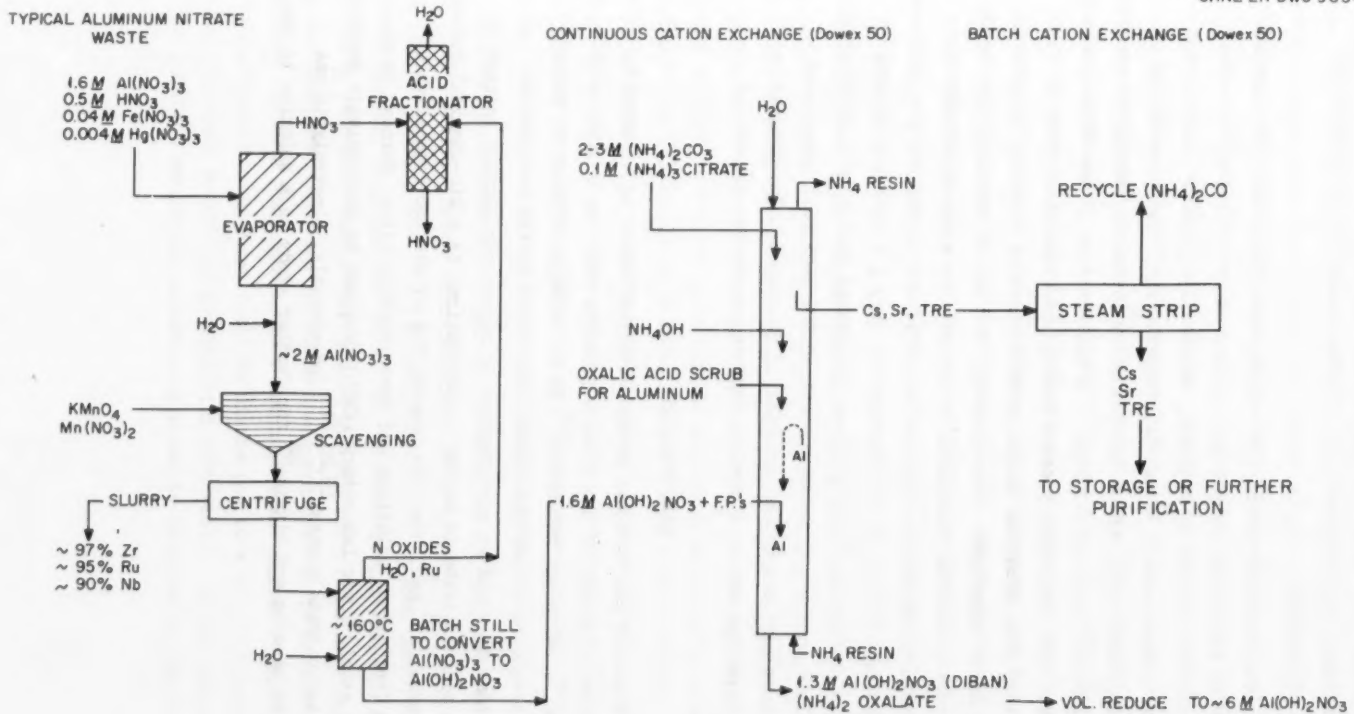

Fig. 4. Removal of Isotopes from Acidic Aluminum Bearing Wastes by the Diban-Ion Exchange Process.

fission products separated for use as isotopes. Alternatively the fission products can be eluted with mineral acids such as nitric or sulfuric, if desired.

Distribution coefficients for cesium, strontium, and rare earths, calculated as radioactivity per unit volume of resin divided by radioactivity per unit volume of solution, between 1.6 M Diban solution and "Dowex 50X8" resin were 7, 8 and 11, respectively. Decontamination of aluminum solutions from cesium and strontium therefore, should be greater than the 10^3-10^4 found previously.[14] A disadvantage of the Diban process is the corrosive conditions present during the evaporation step to form Diban. Since some aluminum wastes contain ammonium nitrate, their evaporation could be hazardous. Consequently, the use of hydroxyl form anion exchange resins will be considered to convert the aluminum nitrate to Diban. Resin decomposition from radiation is not considered a significant factor. Using a figure of 20% degradation when 1 g of resin absorbs 1 watt-hr of radiation,[16] it has been calculated that only 0.0056% of the resin will be lost per cycle when enriched 10% U-Al fuel irradiated to 4×10^{13} n/cm^2/sec and cooled 5 months is processed. The Higgins ion exchange contactor would be used to achieve continuous processing.

ELECTROLYSIS

The alkaline ion exchange method described above is expected to remove essentially all of the fission products with the exception of ruthenium^{106}. Although ruthenium^{106} is not a major biological hazard, it is desirable to reduce the amount discharged to the environment. At the same time it would be advantageous to reduce the nitrate content of the waste since nitrate in higher concentrations is a bio-chemical poison. Early workers[17,18] had shown the feasibility of electro-deposition of ruthenium from dilute solutions and more recently Alter, Barney, et al., at Knolls Atomic Power Laboratory (KAPL) proposed an electrolytic process for treating nitrate wastes.[19,20,21] Consequently, electrolysis was selected by Messing and Higgins[22] for further study as a solution to the problem.

Messing and Higgins considered two types of cells. The first was the KAPL nitrate reduction type which uses a simple stainless steel compartment with close spaced stainless steel cathodes and anodes (Fig. 5). Ruthenium electrodeposits on the cathode and nitrate decomposes to ammonia gas at the cathode. The sodium is converted to caustic which can be reused to neutralize more waste. The second type was a cell designed by Ionics, Inc. which contains cation and anion ion exchange membranes as barriers (Fig. 6).[23] In this cell, the cationic fraction of the ruthenium migrates to and deposits on the cathode. Similarly the sodium and nitrate ions migrate to the cathodic and anodic compartments to form caustic and nitric acid. After a series of tests, the KAPL design was selected for intensive study since it was the simplest and accomplished the objectives of ruthenium and nitrate removal more directly. This type of cell was used in the cost estimate outlined in the preceding section for treating ORNL wastes.[13]

In laboratory scale studies on the nitrate reduction type cell, Messing and Higgins showed that synthetic alkaline waste containing 0.55 - 4.83 M total electrolyte as $NaNO_3$ and NaOH could be decontaminated from ruthenium by factors of >210. The nitrate of 3.0 M total electrolyte waste was reduced to ammonia with current efficiencies of 100%. Power consumption was 4.7 kwh/lb of nitrate reduced. Significant factors affecting the rate of ruthenium decontamination were temperature, cathode area, cathode current density, and electrolyte concentration. Those affecting nitrate reduction current efficiencies were the cathode current density, electrolyte concentration, and stirring rate.

They estimated that a 250-gal cell containing six type-347 stainless steel anodes and five Armco iron cathodes, 48 by 12 in., would process 1,000 liters of 2.0 M $NaNO_3$--0.7 M NaOH waste in 24 hr when operated at a current density of 3 amp/in.^{2} at 100°C. Nitrate levels would be reduced below 0.1 M with decontamination factors of >200 for ruthenium. This is more than adequate to reduce the ruthenium concentration to drinking water tolerances for both ORNL intermediate and high level wastes (Table 1). The sodium hydroxide produced could be discharged to a pit or concentrated by evaporation and reused for neutralizing more waste. In the latter case, only the purified evaporator concentrate would be released to the environment (Fig. 2).

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Table 1. Decontamination Factors Required to Reduce Neutralized Oak Ridge National Laboratory Waste Supernates to MPC_w Values*

	Present ORNL Neutralized Waste Supernate		ORNL Neutralized Waste Supernate Expected from Power Reactor Fuels**		MPC_w
	Specific Activity (μc/ml)	Decontamination Factor	Specific Activity (μc/ml)	Decontamination Factor	(μc/ml)
Sr^{90}	0.1	1.2×10^5	76.0	9.5×10^7	8.0×10^{-7}
Cs^{137}	2.0	1.3×10^4	7.60	5.1×10^6	1.5×10^{-3}
Ru^{106}	0.47	5.7	13.0	130	0.1
TRE(Ce^{144})***	0.40	1.0×10^3	7.6	1.9	4.0×10^{-2}
Pu^{239}	-	-	0.013	8.9×10^3	1.5×10^{-6}

(Table prepared by J. O. Blomeke, ORNL.)

*MPC_w = Maximum permissible concentration in drinking water for life-time use from Report of National Committee on Radiation Protection, National Bureau of Standards Handbook No. 52, (in press).

**Calculated using following assumptions: neutralized waste from fuel enriched to 2% U^{235}; irradiated to 10,000 Mwd/ton; volume = 1.1 gal/kg U; decayed 10 yr; alkaline supernatant will contain 1% of original Sr^{90}, 1% of TRE, 10% of Ru^{106}, and 100% of Cs^{137}; reprocessing losses for $Pu^{239,240}$ were 1% and supernatant contains 1% of process losses.

***Rare earth of lowest MPC_w.

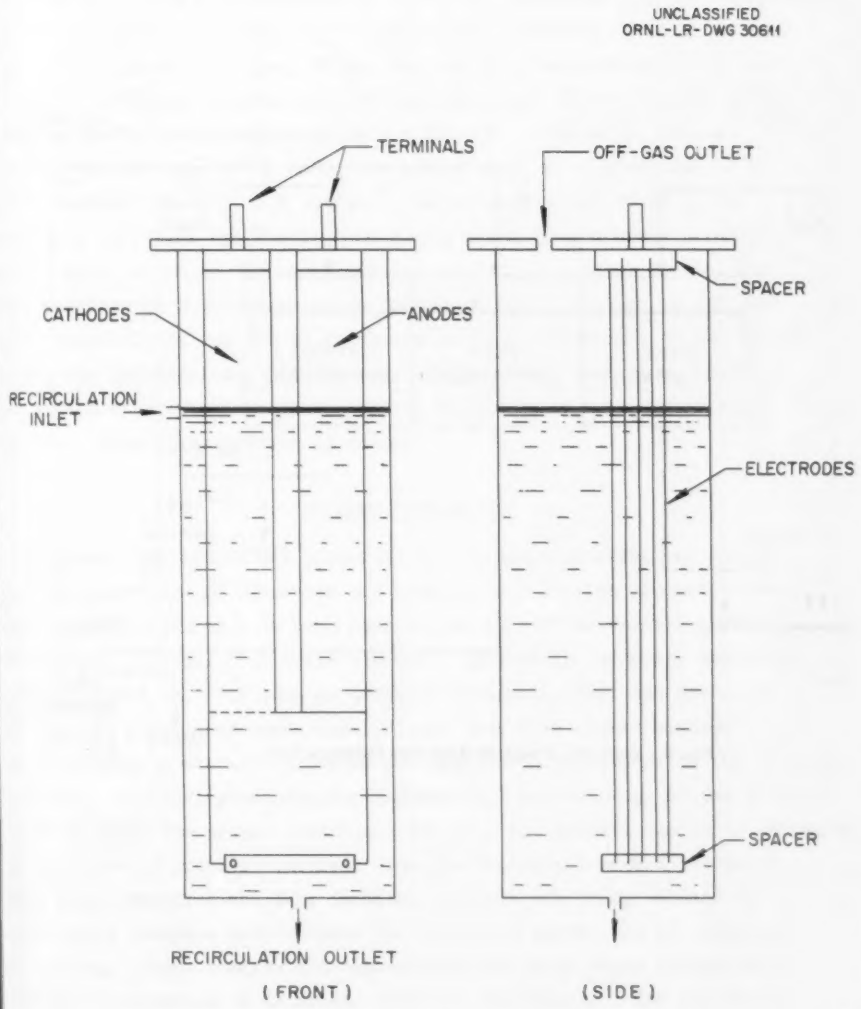

Fig. 5. Nitrate Reduction and Ruthenium Removal Cell.

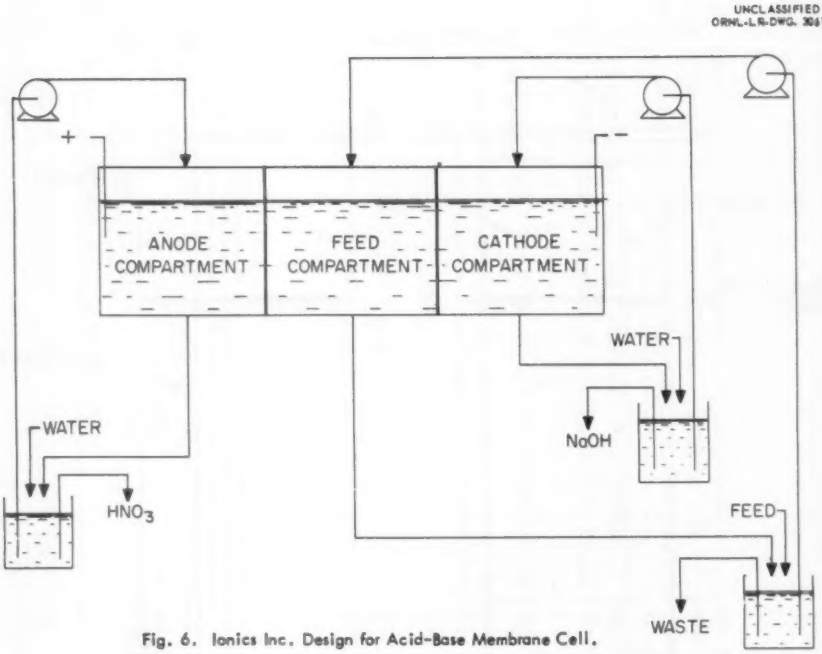

Fig. 6. Ionics Inc. Design for Acid-Base Membrane Cell.

An experiment was run to determine the maximum ruthenium decontamination obtainable. A solution containing 0.44 M $NaNO_3$ and 0.11 M NaOH and Ru-106 tracer, was electrolyzed at a potential of 3 volts, a current density of 0.13 amp/in.^{2}, and a temperature of 60°C. The half-life for the ruthenium deposition was 3.4 hr (Fig. 7). A sharp break occurred at 25 hr in the plot of log gross β vs time, which represented a decontamination factor of 250. Continued electrolysis for an additional 46 hr resulted in an increase in the decontamination factor to 430. Similarly, from a solution containing 3.43 M $NaNO_3$ and 1.40 M NaOH, at a potential of 6.2 volts, current density of 4 amp/in.^{2}, and a temperature of 89°C, the ruthenium half-life was 2.0 hr. As in the above case a break occurred in the plot, at about the same level of activity but after about 15 hr. This represented a decontamination factor of 210. Whether or not the remaining activity was due to ruthenium or to a contaminant of the Ru-106 tracer was undetermined. If the remaining activity was not ruthenium, the ultimate decontamination factor for ruthenium is much higher than 200-400. More investigation is needed.

SOLVENT EXTRACTION

Solvent extraction has proven to be a singularly efficient method for the separation of plutonium and uranium from fission products. Consequently, efforts have been made to use solvent extraction for the recovery of isotopes from waste streams. Inherently, however, the well-known low cost solvents such as tributyl phosphate (TBP) and hexone do not extract fission products easily since they were chosen because of their ability to extract plutonium and uranium in preference to the fission products. Tributyl phosphate, for example, will extract many of the fission products under the proper conditions but will not extract cesium or strontium, the isotopes of greatest interest from the biological hazard viewpoint. Other more expensive and less familiar solvents now being studied in development programs show promise for selective extraction of strontium and cesium. These studies are approaching the point where larger scale testing is warranted to determine over-all feasibility. The following sections present the status of solvent extraction for isotope recovery.

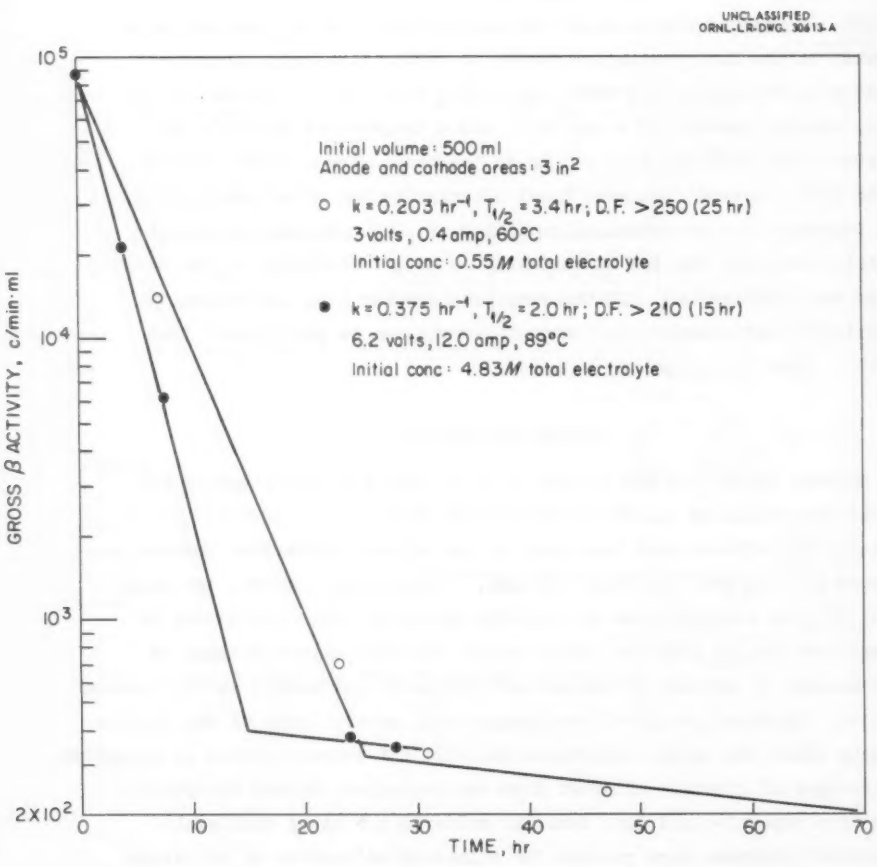

Fig. 7. Demonstration of Maximum Decontamination Factors by Ruthenium Plating.

Organophosphorus Acids. A number of organophosphoric acids have been studied by Coleman, et al., for the selective extraction of strontium and cesium from neutralized wastes.[24,25] Particular emphasis was placed on their separation from sodium which is the bulk ion in neutralized waste. Sodium salts of organophosphorus acids in hydrocarbon solution extracted strontium from sodium nitrate solutions over a wide pH range. The lower limit, the pH at which each extractant was hydrolyzed completely to the hydrogen form, was a function of its acid strength, and the upper limit was set by the phase stability of the organic salt solution. With di(2-ethylhexyl) phosphate (D2EHP) the useful pH range is from about 3 to above 12. The strontium extraction coefficient from 0.5 M sodium nitrate solution with 0.1 M Na-D2EHP rose sharply to a maximum of ~200 near pH 5 (Fig. 8). The separation factor for strontium from sodium, $SF_{Na}^{Sr} = E_a^o(Sr)/E_a^o(Na)$ was about 2000 at the maximum. At this pH the reagent is approximately half in the sodium form, so that the extraction coefficient for sodium, $E_a^o(Na)$ was fixed at 0.05/.5 = 0.1. This separation factor and extraction coefficient should provide an excellent separation of strontium from sodium nitrate solutions.

An extraction isotherm at pH 5.1 leveled off at an Sr/D2EHP mole ratio of 1/4. The shapes of the extraction-vs-pH curve and of the extraction isotherm are attributed to formation of a chelate complex with two D2EHP dimers,

$$H(D2EHP)_2Sr(D2EHP)_2H$$

analogous to the complex found in uranyl extraction with D2EHPA.[26] Thus, the formation of the chelate complex is highest at a pH near 5 where the concentrations of both ionized and un-ionized organic acid are favorable. At lower pH the direct competition by hydrogen ion rapidly increases, while at higher pH ionization increases until only the simple salt (D2EHP)Sr(D2EHP) can be formed. Above pH 7 the strontium extraction coefficient was constant at about 10 (Fig. 8). Here the extraction varied nearly linearly with reagent concentration and with the reciprocal of the aqueous sodium concentration, consistent with existence of the organic salt as a colloidal dispersion in the hydrocarbon solution. The separation factor for strontium from sodium was constant at about 50 over the pH range 7-12. In this case the distribution coefficient for sodium, $E_a^o(Na)$, was

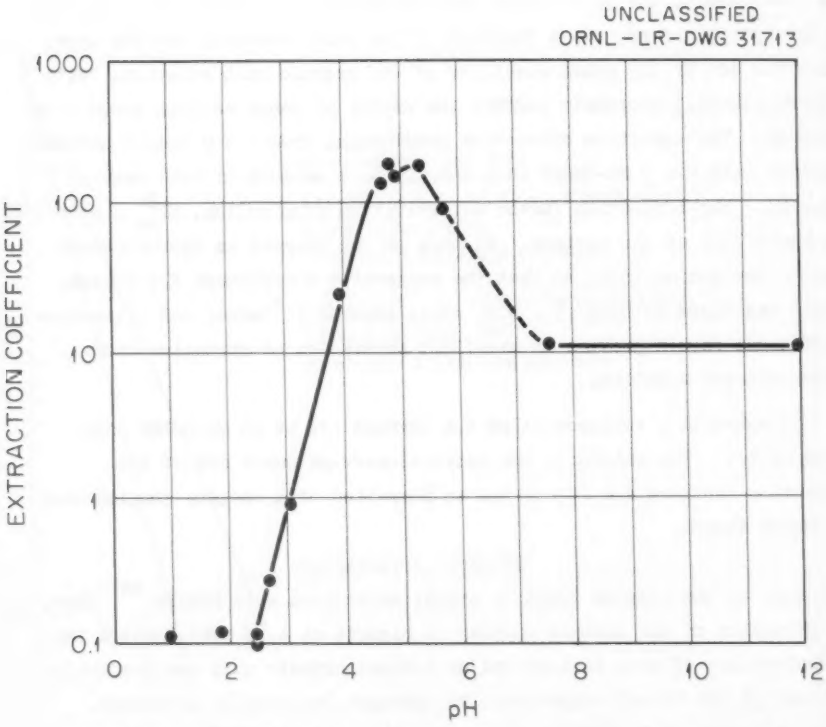

Fig. 8. Strontium Extraction Coefficient vs Aqueous pH. Extracted from ~0.5 M $NaNO_3$ aqueous solution by 0.1 M sodium di(2-ethylhexyl)phosphate in Amsco 125-82 modified with 5% TBP.

fixed at 0.1/0.5 = .2 by the macro concentrations.

Table 2 shows a list of other organic phosphorus acids tested. Only dicyclohexylphosphinic acid failed to extract strontium (i.e., its distribution coefficient was $E_a^o(Sr)$ = 0.1 at pH 3 to 12). None gave higher separations of strontium from sodium than did D2EHP. Several extracted strontium at significantly lower pH than did D2EHP, or gave somewhat higher extraction from alkaline solution.

Table 2. Organophosphorus Acids Tested for Extraction of Strontium[28]

Di-n-octylphosphoric Acid in Amsco 125-82
Bis(diisobutylmethly)phosphoric Acid in Amsco 125-82 + 3% TBP
2, 6, 8-trimethylnonyl-4-phosphoric Acid in Kerosene
2-methyl-7-ethylundecyl-4-phosphoric Acid in Kerosene
3, 9-diethyltridecyl-6-phosphoric Acid in Kerosene
Di-(2-ethylhexyl)phosphinic Acid in Amsco 125-82 + 10% TBP
Di-cyclohexylphosphinic Acid in Benzene
Di-n-hexylphosphinic Acid in Amsco 125-82 + 5% TBP
Phenyl(1-hydroxy-2-ethylhexyl)phosphinic Acid in Benzene

The organophosphorus acids were less successful in extracting cesium than was the case with strontium. Although monoalkylphosphoric acids are not significantly stronger acids than the corresponding dialkylphosphoric acids,[27] the monoalkyl acids in hydrocarbon solution extracted alkali metal ions from aqueous solutions that were more acidic, down to pH 1. The order of selectivity for the alkali metals from nitrate solution with 1-(3-ethylpentyl)-4-ethyloctylphosphoric acid (HDPA) and others of similar structure was Cs > K > Na > Li. The cesium extraction coefficient from 0.5 M sodium nitrate solution with 0.5 M HDPA in kerosene was ~3 in the pH range 1-1.5, with separation factors from sodium of ~30. Cesium extraction by sodium di(2-ethylhexyl)-phosphate was low, with cesium extraction coefficients less than those of sodium in the pH range from 4 to 11.

Cesium was removed from nitrate solution more effectively by sorption on a precipitate of zirconium phytate (inositol hexaphosphate). Sorption was highest from slightly acidic solutions, but still considerable from 2 M nitric acid in single-stage batch equilibrations using 1 g of sorbent per 100 ml of solution (Table 3).

Table 3. Cesium Sorption from Nitrate Solutions by Solid Zirconium Phytate[28]

Aqueous Solution	pH	Per Cent of Cs Sorbed
0.5 M $NaNO_3$	12	6
0.5 M $NaNO_3$	0.7 to 2.2	80
2 M HNO_3	0	35
1 M $Al(NO_3)_3$	1	97
2 M $Al(NO_3)_3$	1	92

Equilibrium was reached in 5 min. The limiting loading from 0.1 M cesium nitrate solution was about 1 mmole of cesium per gram of sorbent, and the limiting loading from 0.5 M $NaNO_3$ containing 10 ppm cesium at pH 3 was 2 mg of cesium per gram of sorbent. A column sorbed 0.7 mg of cesium per gram of sorbent from 1 M 10 $NaNO_3$ solution containing 10 ppm cesium at pH 1 before breakthrough. The sorbed cesium can be eluted with ammonium nitrate or chloride.

Tributyl Phosphate (TBP). In the absence of significant quantities of TBP-extractables, such as uranium or thorium nitrates, which create TBP-saturation and/or strong back-salting effects on less extractable species the cerium and rare earths are readily extracted by tributylphosphate. In cases where nitric acid is the only aqueous salting agent, relatively strong concentrations of acid e.g., 8-16 M are required to provide usefully high rare earth distribution coefficients because the TBP-extractable nitric acid may effectively back-salt the rare earth nitrates. However, such systems have been effectively employed in the extraction and separation of the naturally occurring rare earth elements by Weaver and Topp.[29] In aqueous systems containing aluminum nitrate and under either acid-deficient or

relatively low acidity conditions, fairly low salt-strengths procure usable distribution coefficients for the cerium and yttrium rare earths. Both pure TBP and 30-45% solution of TBP in inert solvents have been studied.

In tracer studies employing 1.0 volume of synthetic solution (1.6 M $Al(NO_3)_3$, 0.16 M HNO_3, 0.02 M $SO_4^=$) with 0.2 volume of 100% TBP, Krieger et al.[30] found that a combination of chemical treatments of the aqueous waste provided a system for extraction of the cerium, yttrium, zirconium, niobium and ruthenium fission products. Greater than 99% of the rare earths were extractable without treatment. The addition of cupferron was employed to attain about 89% extraction of zirconium-niobium; and reduction by metal addition to aqueous phase was employed to attain approximately 80% removal of the ruthenium.

In other laboratory studies, wastes from the Thorex process were treated by Gresky and Wischow.[31] The cerium rare earths were extracted from the evaporated waste concentrated with 42% TBP in Amsco diluent. The concentrates contained 2.47 M $Al(NO_3)_3$-0.11 M $SO_4^=$ and were 0.87 M acid deficient. Employing a batch countercurrent extraction and stripping system, it was possible to completely extract and concentrate the cerium rare earths. Extraction distribution coefficients for the latter were observed to be greater than 10^4, making it possible to employ small efficient organic volumes for the extraction process.

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FISSION PRODUCT RECOVERY FROM RADIOACTIVE EFFLUENTS

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INTRODUCTION

Of the many fission product isotopes formed in the fission reaction, some are of sufficiently long half-life and formed in sufficient yield to be of major importance from the standpoint of long-term waste disposal or retention or of recovery for utilization as radiation sources. The more important of these isotopes, with their daughters, are shown in Table I. Cesium-137 is promising as a gamma radiation source becuase of its long half-life, high fission yield, and high gamma/beta ratio. Strontium-90 is of interest for applications where a long-lived beta-only emitter is desired. The others are of lesser interest although they may find application where their short half-lives are not a serious deterrent. In the area of waste disposal or retention, interest centers on the removal of cesium-137 and strontium-90, again because of their long half-lives and because of the severe biological hazard of strontium-90.

TABLE I

RADIATION CHARACTERISTICS OF PRINCIPAL FISSION PRODUCTS

Nuclide	Thermal Fission Yield from U-235, %	Half-Life	Principal Radiation, MEV	
			β	γ
Cs-Ba-137	5.9	30 y	1.2	0.66
Sr-Y-90	5.9	28 y	2.2	None
Ru-Rh-106	0.38	1.0 y	3.53	0.51 (25 %) 2.41 (0.3 %)
Zr-Nb-95	6.4	65 d Zr, 35 d Nb	0.4	0.75
Ce-Pr-144	6.1	285 d	3.0	2.2 (1 %)

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This paper describes some processes, presently under development in the Hanford Laboratories, for the recovery of certain fission products, particularly cesium, from currently available process effluents. The proposed processes are, to some extent, similar to processes previously used successfully at Hanford for the pre-disposal treatment of aged process effluents. Hanford's experiences with this program are described briefly.

Fission product recovery, particularly the removal of long-lived isotopes from process effluents, is frequently considered an integral part of ultimate waste disposal. A common concept is that, if cesium-137 and strontium-90 are removed, the remainder can be disposed of after a relatively short storage period during which short-lived isotopes will decay to non-hazardous levels. Some discussion of the relationship of fission product recovery to ultimate waste disposal and of the limitations of this relationship is presented in the final section of this paper.

FISSION PRODUCT RECOVERY

Research at Hanford on fission product recovery has concentrated largely on cesium since it was felt that this is the fission product most likely to find a large scale market in the near future. The recovery of other fission products has been considered to be of secondary interest, and schemes for their recovery have accordingly been designed to fit into a main-line cesium recovery process. Furthermore, interest centered on the use of precipitation techniques because of the existence at Hanford of idle precipitation facilities.

It should be noted further that the selection of an optimum process for the recovery of fission products from a radioactive waste solution will be very dependent on the composition of the waste and the method of waste disposal employed. Thus Oak Ridge has successfully used the fractional crystallization of ammonium alum to recover cesium from the high aluminum nitrate ARCO chemical separations plant waste[1], and the British are reported to use phosphotungstate carrying to recover cesium from their waste, a nitric acid solution which is stored in stainless steel tanks without neutralization.[2] At Hanford, uranium and plutonium are recovered from irradiated fuel elements by means of the Purex process.[3] This is a solvent extraction process which employs tri-n-butyl phosphate as extractant and nitric acid as salting agent. The bulk of the nitric acid is recovered from the waste by distillation for reuse, and the concentrated waste is then neutralized with caustic and stored in the alkaline condition in underground mild steel tanks. The flowsheet composition of this waste is shown in Table II. In actual practice, the composition of the unneutralized waste is rather variable with the concentration of many of the inert constituents often exceeding the above values and with traces of corrosion products, uranium, and aluminum also present. This is shown by the numbers in the third column. Although large quantities of fission products are present in the Hanford storage tanks, recovery from this material would be more expensive than from the current plant waste, and the latter should be adequate to satisfy the expected demand. Therefore effort was concentrated on recovery of cesium from the fresh waste.

TABLE II

COMPOSITION OF PUREX WASTE

Constituent	Molarity	
	Flowsheet	"Actual"
HNO_3	7.6	6
Fe	0.11	0.3
Al	-	0.1
Na	0.38	1
U, Cr, Ni	Trace	Trace
$SO_4^=$	0.21	0.4
Total Fission Products	~0.01	~0.01
Cesium	~0.001	~0.001

Several precipitating agents were scouted in the initial work. These included tetraphenyl boron, cobaltinitrite, silicotungstate, and various ferri- and ferrocyanides. The latter showed the most promise and were selected for intensive development. As has been found to be the case with many other scavenger reactions, coformed precipitates were found to be much more effective than preformed. The ferrocyanides of zinc, nickel, copper, iron(III), iron(II), cobalt, cadmium, uranium(VI), and manganese(II) and the ferricyanides of nickel, copper, and zinc all precipitated cesium to some degree. From the standpoint of extent of cesium recovery, volume of precipitate to be handled, and specific activity of product, zinc ferrocyanide, zinc ferricyanide, nickel ferrocyanide, and iron(III) ferrocyanide were the most efficient. Nickel ferrocyanide was effective over the widest pH range (from about 5 M nitric acid to pH 10) whereas zinc ferrocyanide gave the smallest precipitate volumes and highest specific activity. Details on the effects of variables are reported elsewhere.(4)

A flowsheet based on the use of zinc ferrocyanide is shown in Figure 1. Noteworthy features of the cesium zinc ferrocyanide process are the large volume reduction (ca. 1000 fold) and the high specific activity of the precipitate, which is about 45 percent cesium by weight (dry basis). The precipitated cesium is also of high radiochemical purity since most of the fission products are removed on the ferric hydroxide precipitate. This process has been tested with full level plant waste and is currently undergoing pilot plant development.

The product of the cesium zinc ferrocyanide process is a definite compound with the composition $Cs_2ZnFe(CN)_6$.(4) It appears to have a lower solubility

than zinc ferrocyanide itself, and most complete cesium precipitation occurs when zinc and ferrocyanide are added in quantities stoichiometric to the cesium. Although cesium zinc ferrocyanide is acceptable from a specific activity standpoint since it is very nearly half cesium by weight, there is some reason to think that it may undergo slow radiation decomposition with resultant gas evolution. Therefore, methods have been developed to convert it to cesium chloride, which is known to be satisfactory for irradiation source use. This can be done in a variety of ways,[4] e.g., by calcination followed by water leaching, acidification of the leach solution with hydrochloric acid, and evaporation to dryness; by dissolution in sulfuric acid followed by sulfide precipitation of iron and zinc, and anion exchange removal of sulfate; or by hydrolysis with steam[5] at 250-300°C followed by leaching, acidification, and drying. The latter process has been selected for engineering prototype testing.

The most difficult step in the cesium zinc ferrocyanide process is the centrifugation (or filtration) and washing of the ferric hydroxide precipitate. The handling characteristics of this precipitate are considerably improved by slow neutralization with gaseous ammonia or by urea hydrolysis (homogenous precipitation); however, the large bulk of the precipitate is still a problem at high iron concentrations. Therefore, the removal of cesium after only partial neutralization and prior to iron precipitation has also been investigated.[6] Nickel ferrocyanide, zinc ferricyanide, and zinc cobalticyanide were the most efficient cesium carriers at low pH, with the zinc ferricyanide giving the best recoveries and smallest precipitate volumes. Recoveries of 85 to 98 percent were obtained from full level plant waste solution. However, it was necessary to use a ten-fold excess of precipitant, and the precipitate was contaminated with up to ten percent of the initial zirconium and ruthenium activities. Thus, further processing would probably be required to improve the radiochemical purity and specific activity of the cesium product. Choice between the "acid side" flowsheet and the cesium zinc ferrocyanide process for a particular application would probably depend on the composition (iron, uranium, and aluminum concentration) of the waste and on the results of larger scale comparative tests.

A scheme for the recovery of other fission products, in addition to cesium, is shown in Figure 2. Control of pH is utilized to fractionate the fission products into groups, and appropriate precipitation reactions are then utilized to isolate individual elements. Final packaging of strontium, cerium, and rare earths would probably be as the fluorides or fused oxides, since these have good chemical and radiation stability. Cesium recovery would be by ferrocyanide precipitation, as before.

An engineering study of the probable cost of producing fission products in Hanford facilities has been made.[7] It appears that cesium-137 (as cesium chloride) can be produced and sold at a price of about 50 cents per curie at a processing rate of the order of a megacurie per year. Production at a high rate or concurrent production of other fission products would reduce this price substantially.

HANFORD WASTE SCAVENGING EXPERIENCE

Procedures similar to those just described for the recovery of cesium and strontium have been used successfully at Hanford for the decontamination of certain process effluents prior to discharge to the ground. The solutions contained ferric, sulfate, and phosphate ions in moderate concentrations (< 0.5 M) in addition to high concentrations of nitric acid and/or sodium nitrate. By simply making the solutions alkaline (pH 8-10) ferric hydroxide and phosphate were precipitated. Most of the total radioactive content present was coprecipitated with these solids. Decontamination factors for strontium ranged from 80 to 200. However, very little of the cesium present was removed. Precipitation of nickel ferrocyanide prior to or during neutralization of acidic wastes or in the supernatant liquid from already alkaline wastes removed cesium very thoroughly. Cesium decontamination factors as high as 10^4 were obtained, and factors in excess of 10^3 were routine for a single strike in large-scale processing. Ferrocyanides other than nickel (copper, iron, cobalt, zinc) are also effective carrier precipitants for cesium as noted elsewhere in this paper. Nickel ferrocyanide was chosen for this application because it is relatively inexpensive and becuase it functioned best in the pH range (8-10) dictated by other aspects of the process. More complete removal of strontium than that obtained by making the solutions alkaline was desired. By adding inert calcium or strontium nitrate to the solutions prior to treatment, strontium decontamination factors were increased to 500 or greater. Following settling of the solids produced in these treatments, the supernatant liquids contained strontium-90 and cesium-137 in the range of 0.02 to 0.1 uc/ml. Total fission product content ranged from one to 20 uc/ml. depending on the age of the effluent. References 8, 9, and 10 describe these procedures in much greater detail.

It should be emphasized that the procedures just described did not produce an effluent which could be discharged directly to public waters. Concentrations of many of the isotopes present were many-fold greater than the maximum permissible in drinking water. These effluents could be discharged to ground safely at Hanford only because of an unusually favorable combination of climate and geology, as well as remoteness from habitation and from public waters. The discharge area has a deep ground water table (250-350 feet) overlaid with a sandy, permeable soil having considerable adsorption capacity for many cations. Ground water under the discharge area has a low hydrologic gradient and moves toward the adjacent Columbia River slowly. At no point in the area is ground water removed for domestic or agricultural purposes. Wastes discharged into the ground near the surface percolate through a long exchange column of soil before reaching ground water. Much of the residual fission product content is absorbed by the soil. Of those radioisotopes which do reach ground water, only those with relatively long half-lives (>3 years) are of prime concern since the short-lived isotopes will decay to sub-hazardous levels before reaching the Columbia River. Thus, if the discharge system is so operated that no long-lived isotopes reach ground water in hazardous concentrations, the operation as a whole is safe. In practice, the operating criterion adopted was that no radioactive nuclide having a half-life greater than three years should exist in the ground water at concentrations greater than 0.1 the maximum permissible in potable water. Several million gallons of separations process effluents were treated and

discharged to ground in this program. Total cost including preparation of the discharge sites was about five cents per gallon of waste treated.

Cesium-137 and strontium-90 are the only fission products having long half-life and produced in high yield in the neutron irradiation of uranium. Low concentrations of long-lived alpha emitting isotopes were also present in the scavenged waste. However, past experience has shown that these latter are much more completely adsorbed and strongly held in soil than cesium or strontium. Consequently, the quantity of waste which could be discharged to any given area of soil was limited by the adsorption of cesium-137 and strontium-90. Since maximum permissible concentrations in potable water (MPC) for cesium-137 and strontium-90 are 1.5×10^{-3} and 8×10^{-7} uc/ml, respectively,[11] it is apparent that operation of the discharge system within the established criterion required that additional decontamination factors of about 10^3 for cesium-137 and 10^6 for strontium-90 be obtained through adsorption of these isotopes by the soil.

To determine approximately how much of the scavenged waste could be discharged per unit discharge area, soil column tests were made on each batch (300,000 to 500,000 gallons). In these, the number of column volumes of waste passed through the soil column before either cesium-137 or strontium-90 in the effluent exceeded 0.1 MPC was determined. This value and an estimated effective soil column length to ground water were used to establish the volume of waste which could be discharged to any given area.

The testing program brought to light some interesting observations on the effect of various constituents on the adsorption of strontium and cesium by soil. For example, it was found that the adsorption of strontium depended on the amount of phosphate present,[12,13] and a minimum phosphate concentration in the discharged waste had to be specified. Also, it was found that very low concentrations of aluminum (in the range of 0.001 M) had a seriously adverse affect on the adsorption of cesium and strontium. Although not a fission product, cobalt-60 was one of the more troublesome isotopes in this disposal program. It was present in low concentration, presumably due to neutron reactions with impurities in fuel or cladding materials. For some reason, as yet not completely explained, adsorption of cobalt from the scavenged wastes by soil was essentially negligible. Consequently, it was necessary that the cobalt-60 content in the discharged waste be no higher than that specified for ground water. Since cobalt-60 has an intermediate half-life (5.2 years) this specification was set at the MPC or 4×10^{-4} uc/ml for some waste batches. The cobalt-60 which does reach ground water will be used as a tracer for further determinations of ground water flow. These effects imposed very severe limitations on the allowable volume discharged per unit area for some batches of waste. They are cited here to emphasize the caution required in carrying out a waste disposal program even under exceptionally favorable conditions. Under less favorable conditions the problems would be increased many fold.

RELATION OF FISSION PRODUCT RECOVERY TO WASTE DISPOSAL

Historically the recovery of certain fission products, particularly long-lived nuclides, from separations waste has become identified as an integral

part of ultimate waste disposal. In particular, it is argued that, if the long-lived isotopes such as cesium and strontium are removed, the remainder can, after a few years storage, be relatively easily disposed. It would be well to examine this concept in terms of the objectives of a fission product recovery program. The current incentive for the recovery of significantly large amounts of fission products presages use of their radiant energy for processing of foods, destruction of parasites, and initiation of chemical reactions. Economic studies have shown that recovered fission products must be made available at very low cost to compete with conventional forms of energy where these will perform the same task. Even where the effect desired can be obtained only by high energy radiation, fission products must compete economically with machine-made radiation. Thus, the objectives of a large-scale fission product recovery program are the long-familiar ones of low cost and high volume.

Separations process effluents currently available for large-scale fission product recovery contain cesium-137 and strontium-90 in concentrations of between 10^3 and 10^4 uc/ml. To reduce these nuclides to drinking water tolerances, minimum decontamination factors of about 10^6 and 10^9, respectively, would be required. Suggested recovery processes give decontamination factors ranging from only about ten to 100. This is due to the necessity of keeping the process simple and producing a product of high specific activity. Economically competitive processes giving decontamination factors of 10^3 may possibly be devised, but it appears highly improbable that competitive processes giving decontamination factors of 10^6 to 10^9 can be developed. Effluents from presently suggested cesium and strontium recovery processes will contain many times drinking water tolerances for cesium-137 and strontium-90. Thus, even though these effluents are stored until all short-lived nuclides have decayed to sub-hazardous levels, they still cannot be safely discharged where they might contaminate public waters. Complete confinement of the wastes, further treatment to reduce these isotopes to non-hazardous levels, or controlled disposal to ground under favorable conditons of soil retention and remoteness from public waters would be required. It is well to remember that process effluents from which fission products may be recovered also contain small amounts of plutonium, uranium, and other alpha emitting isotopes. These have generally much longer half-lives than and represent about as great a hazard as most fission products. Their presence must be considered in the disposal of any effluent where it may enter public waters.

There are however, undeniable fringe benefits to waste disposal resulting from fission product recovery. Thus, the magnitude of the hazard or catastrophe should a waste storage tank rupture would certainly be diminished if most of the cesium and strontium had been removed. Also,the energy produced by radioactive decay is adsorbed by surrounding media and converted into thermal energy. In highly radioactive solutions, the decay heat may be sufficient to produce boiling while in large masses of highly radioactive solids (as would be derived by evaporating radioactive wastes to dryness) very high temperatures may be reached if thermal conductivity is low. Long-lived isotopes account for a relatively small portion of the decay energy of short-cooled wastes from the processing of irradiated uranium. The Cs^{137} -Ba^{137} and Sr^{90} -Y^{90} parent-daughter chains together account for less than ten percent of the total decay energy in process effluents at usual processing

times (about 100 days after removal from a reactor). However, after five years ageing of the wastes, they account for more than two thirds of the total decay energy. In long-term storage or ultimate disposal techniques in which decay heat is a problem, prior removal of cesium-137 and strontium-90 will reduce the problem. It has been calculated that, under some loading conditions, underground waste storage tanks at Hanford might boil (requiring cooling or condensate return) for a century or more. Removal of most of the cesium and strontium prior to storage of the wastes would reduce this time to a few years.

In summary, recovery of specific fission products from process effluents may reduce heating problems in long term retention of radioactive wastes. It appears unlikely that the very high cesium-137 and strontium-90 decontamination factors required to permit eventual uncontrolled release of the wastes will be obtained in economically competitive recovery programs.

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CESIUM RECOVERY FROM PUREX WASTE
CESIUM ZINC FERROCYANIDE PROCESS

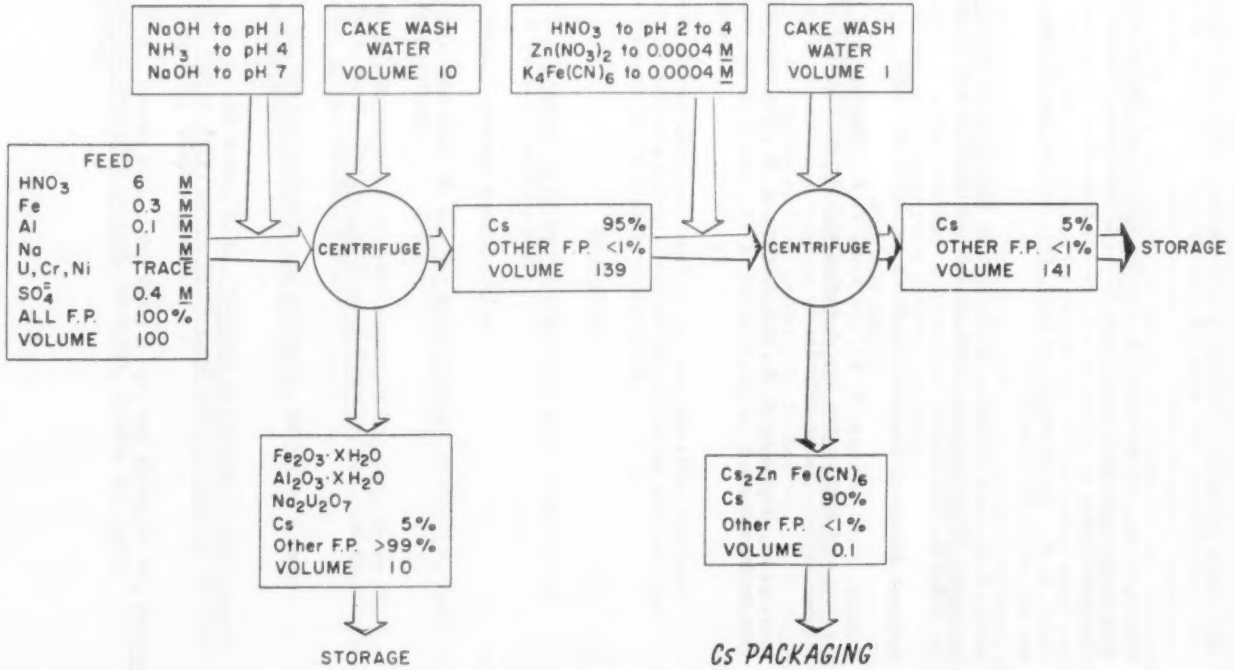

FIGURE 1

FIGURE 1

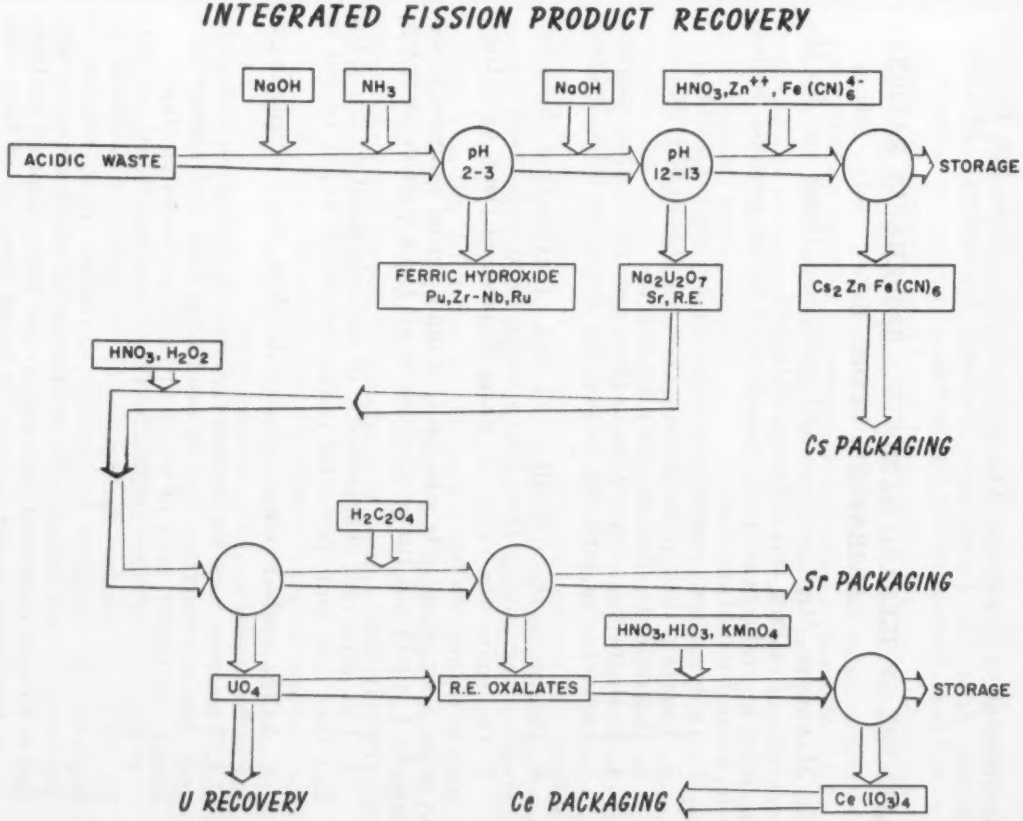

FIGURE 2

Representative HOLIFIELD. The next witness will be Mr. Bernard Manowitz, from the Brookhaven National Laboratory. His testimony will be on fission product utilization.

STATEMENT OF BERNARD MANOWITZ,[8] BROOKHAVEN NATIONAL LABORATORY, UPTON, N.Y.

Mr. MANOWITZ. Mr. Chairman and gentlemen, thank you for the opportunity of making this summary statement.

In order to evaluate properly the effect of fission product utilization on waste disposal one must—

1. Analyze the characteristics of the radiochemical wastes;
2. Ascertain their probable supply as a function of time;
3. Determine to what uses they can be put;
4. Evaluate the competition to these uses from other sources;
5. Estimate the probable market for the extractable byproducts;
6. Determine the difficulty and cost of extracting these byproducts and putting them into a useful form, and finally;
7. Determine the effect on waste disposal if these operations were to be undertaken.

In order to fit potentially into the 15-minute period assigned to me, I thought I would put the conclusions to all those statements immediately, namely that:

1. The important characteristic of radiochemical wastes is the fact that they emit powerful radiations which might be put to use;
2. As the nuclear power industry develops, their supply is going to be ample;
3. Numerous uses for intense radiation in initiating biological and chemical reactions and in producing heat and energy have been investigated, most of which are still in a research stage;
4. Each one of these applications can be accomplished, not only by the fission product radiation, but also radiation produced by high-voltage machines by nuclear radiations from reactors or reactor-produced isotopes, the production of which requires more fission to take place and thus add to the waste disposal problem, or, in most cases, more cheaply by using chemical or heat techniques rather than radiation;
5. Although the market for these byproducts in the immediate future is small, efforts are being made to develop a market;
6. The difficulty of extracting byproducts and putting them into a useful form is considerable, though probably accomplishable at a reasonable cost; and, finally,

[8] Born in 1922 in Jersey City, N.J., has an M.S. degree in chemical engineering from Columbia University. From 1944 to 1947 he worked at Clinton Laboratories, now Oak Ridge National Laboratory, primarily in the field of pile engineering. From 1947 to the present he has been at Brookhaven National Laboratory. There his responsibilities have included the design and construction of the Brookhaven waste disposal system, engineering research on waste processing methods, fission product utilization studies, and research and development work in radiation physics and radiation chemistry. He is now the head of the Radiochemical Technology Group at Brookhaven. From 1952 to 1954 he was a member of the AEC Waste Processing Committee. He was also an adviser to the delegation at the 1955 Geneva Conference where he presented a U.S. paper on waste disposal. Mr. Manowitz is author of a considerable number of publications both in the field of waste disposal and in the field of applied radiation. At the present time he is also an editor of the International Journal of Applied Radiation and Isotopes.

7. Even if fission product utilization is highly successful and widely put into practice, it will probably have a relatively small effect on the waste disposal problem.

These conclusions should not be construed as an indication that applied radiation has a poor future, or that fission byproducts have no market value. On the contrary, I believe that applications of intense radiation have an important contribution to make to the future economy of this country, and that appreciable revenue is possible from the sale of fission byproducts. However, I feel that the growth of this new field is unlikely to provide a solution to the problem of radiochemical waste disposal.

To develop the argument, table 1 lists characteristics of the important long-lived nuclides in radiochemical wastes. I believe these are probably the same radionuclides Mr. Tomlinson described to you a moment before. Note that there are a large number of radionuclides present which belong to different chemical families. Of these, several, i.e., promethium 147, ruthenium 106, and cerium 144, will decay to negligible levels in about 20 years; cesium 137 and strontium 90 will persist for generations, and samarium 151, americium 241, and plutonium 239 will endure for centuries.

Note also the difference in the half lives. The half lives range between a year or so to 25 or 30 years for cesium and strontium; to 24,000 years for plutonium. So some of these will decay in tens of years, others will persist for hundreds of years, and others will persist for thousands of years.

TABLE 1.—*Important characteristics of long-lived nuclides*

Nuclide	Half-life years	Potential supply from power reactor program in—				Extraction efficiency required before discharge of typical wastes to environment after 10 years storage, percent
		5 years		25 years		
		Megacuries	Megawatts	Megacuries	Megawatts	
Cs^{137}	30.0	40	0.2	2000	9	> 99.99999
Sr^{90}	28.0	40	.2	2000	9	> 99.999999
Pm^{147}	2.6	100	.05	3000	1.2	> 99.99999
Sm^{151}	80.0	1	.0001	50	.008	> 99.99
Am^{241}	470.0					> 99.999
Ru^{106}	1.0	20	.2	500	4	> 99.999999
Ce^{144}	0.8	300	2	6000	42	> 99.999999
Pu^{239}	24,000.0					> 99.999
Kr^{85}	10.0	4	.006	200	.2	
Total			2.7		65	

Also in table 1 is an estimate of the potential supply of these radionuclides as a result of the U.S. power reactor program based on the Palmer Putnam prediction of the growth of the U.S. nuclear economy. The data indicate that in terms of curies, enormous amounts of these radionuclides will accumulate. In terms also of the total amount of heat which can be generated from these radionuclides an appreciable amount is also going to accumulate. An estimate of the market for these fission products in the immediate future is very difficult to make because so many items, such as cost and usefulness, are still unknown. I think it can be stated categorically that in every case the potential supply far exceeds the demand that can be foreseen in the immediate

future. However, the Office of Isotope Development of the Atomic Energy Commission has recently initiated a program designed to accelerate the uses of radioisotopes, and to develop an industrial market. This program includes:

1. Radioisotope applications contracts to develop new, or to improve existing practical uses of radioisotopes;
2. Industrial radiation contracts to accelerate development of massive radiation sources and technology for economic applications of radiation of high power output;
3. Isotope production activities to make available commercial quantities of radioisotopes at economical prices, and to encourage their production by industry;
4. Increased opportunities to industrial personnel for training in the technology necessary for practical application of isotopes.

The amounts of radioactivity needed for tracers, or thickness gages, or instrumentation, are very small, even for a greatly expanded market. The only hope for making an appreciable dent in the potential supply of radionuclides is by the use of massive radiation sources. It is highly probable that if massive radiation sources are marketed, for safety reasons they will have to be sealed sources. A sealed source must be radiation stable—that is, it must not evolve gases which might pressurize the container.

It is unlikely that mixed fission products could be put into a chemical form that would be sufficiently radiation stable. Furthermore, mixed fission products would have a complex decay pattern, making the planning of precise dose rates and total irradiation extremely complicated. It is highly likely, therefore, that individual radionuclides would have to be separated and properly packaged to make up massive radiation sources. Cesium 137 is the most likely fission product to be useful as a gamma source. Some potential applications for massive amounts of gamma radiation are the following:

1. Radiography and therapy: Cesium 137 is and will continue to be competitive with high voltage machines, and cobalt 60 for the radiography and therapy markets.

The total potential market, however, is very small compared with the potential supply of cesium 137.

2. Sterilization of drugs and medical supplies: The sterilization of drugs and medical supplies will probably provide a relatively small radiation market. Chemical and steam techniques in use today are cheap and in most instances entirely adequate.

For those uses where radiation is applicable, high voltage machines, since they can be developed faster, will probably capture an appreciable fraction of the market. Cobalt 60 will be competitive and probably preferable to cesium 137 in those instances where deep penetration is necessary, since cobalt 60 has more penetrating radiations.

3. Sterilization and pasteurization of food: Should the technical difficulties of storing foods for long periods of time without refrigeration by radiation treatment be surmounted, a very large market for many types of radiation sources could develop. At the moment, the Quartermaster Corps program on food sterilization has had the most research and development attention, but the program is not yet at a point where a process and process equipment can be defined. Therefore, a market cannot as yet be estimated. It is my understanding that

in about 1½ to 2 years process and process equipment information will be available.

Not as much work has been done on food pasteurization as on food sterilization, so that the commercial market for radiation preserved foods can be even less well defined. Even if large potential markets exist, cesium 137 radiation sources will face stiff competition from high voltage machines, cobalt 60 and sodium 24 reactor coolants.

Studies have recently been made at Brookhaven comparing cesium 137, cobalt 60, and sodium 24 as sources for a high intensity food irradiator similar to the proposed Quartermaster Corps HIFI. For the case of cesium 137 it would be necessary to develop techniques to prepare the cesium 137 to at least twice the specific activity it can be prepared in today. Even at that level it would require about 30 million curies, or approximately the total estimated amount that would be accumulated from the entire reactor program in the next 5 years, in order to provide enough activity for a single 3,000-pound-per-hour irradiator. On the other hand, the Quartermaster Corps HIFI will use about 2 million curies of cobalt 60 at a specific activity readily available. Even less sodium 24 would be needed, although the required specific activity would involve changes in existing sodium-cooled power reactor design.

4. Initiating chemical reactions: A wide variety of chemical reactions that can be effected by radiation have been and are being studied, ranging from rubber vulcanization and petroleum cracking to the making of specialty plastics. Most of this work is still in an embryonic stage so that a market cannot be well defined. Once again, even if a large market can be developed, cesium 137 will hardly be competitive with high voltage machines, cobalt 60 or sodium 24. In this case, nuclear reactors themselves may very well be used as the radiation source, generating rather than reducing waste volumes. In this area particularly, chemical and heat techniques for making the same or a similar product will be difficult for radiation to compete against. Potentially, cesium 137 can also be used as a heat source. As table 1 indicates, eventually there will be a sufficient supply of cesium 137 so that power supplies in the region of hundreds and perhaps thousands of kilowatts can be considered. Here the unique properties of cesium 137, that is, its half life, and the fact that it can be packaged in small volume, may make it especially valuable for a heat and power supply in remote places.

Strontium 90 and prometheum 147 are pure beta emitters and can be considered as materials for massive beta sources. Krypton 85 is also beta emitter and can be considered for specialized uses.

Massive beta sources can be used for surface sterilization and surface pasteurization of foods and other items. Potentially, they could be used also for the initiation of reactions in the gas phase and for the treatment of materials in thin sheet or film form.

For these uses strontium 90 and prometheum 147 would have to compete against high voltage machines. Krypton 85 can be used in the manufacture of light sources, such as railroad markers and uses of that kind. For this use it would have to compete against tritium which is a reactor produced material. Prometheum 147 is particularly applicable for atomic batteries.

Strontium 90 may also ultimately be useful as a specialized heat and power source.

Further information on potential markets is being developed for the Atomic Energy Commission by the CEM group of Washington, D.C., by A. D. Little of Cambridge, Mass., and by Radiation Applications, Inc., of New York City.

I would summarize my feelings on potential markets for fission products something like this.

Normally, when one is developing a standard commercial product, as, for example, a new plastic, it takes about 7 years to get from test tube to a marketed product. During this interval one has accumulated basic information on the feasibility of the process, engineering information on the equipment required to effect the process, pilot plant information on the practicality of the process, information on the properties, quality, and acceptability of the product, market analysis information, and has constructed and operated a plant for producing the product. In this normal chain of events there is already available to the engineer and to the scientist a vast amount of basic information on the characteristics of the form of energy used in the process, and on the characteristics of many types of equipment through which energy can be infused into the process.

For example, if you are designing a heat exchanger you can go to the handbooks and get much information on heat transfer coefficients and information of that sort.

The situation is not the same when one considers radiation as a source of energy. Much basic information has yet to be developed on the interaction of radiation with matter, and on the characteristics of various types of radiation sources. This can be achieved only by sustained basic engineering research which, in this particular field, requires complex and expensive facilities, as, for example, the new type of source preparation cells and source evaluation cells that we are proposing at Brookhaven. One still needs a handbook on radiation source design and on radiation effects containing information determined empirically and substantiated at full activity level. We are hoping to develop this basic information at Brookhaven within the next several years. Even after this information has been accumulated, development work on radiation processes will be slower and probably more expensive than on processes where one uses only heat or chemicals to product a product.

The present status of the applied radiation field is that most applications have had at the most 3 or 4 years of basic research, in all instances using laboratory radiation sources that bear no resemblance to sources that would be optimum for a commercial plant. Thus, it is still too early to make any realistic evaluation of potential markets, but if planned programs are actually carried out, we should be in a much better position to make such a realistic evaluation in 3 or 4 years.

The difficulty in extracting specific fission products and putting them in a usable form will be described in detail by Mr. Art Rupp, of ORNL, and have previously been described by Mr. Tomlinson. Suffice it to say that in my opinion Mr. Rupp and his coworkers have done an outstanding job in developing techniques for preparing radioisotopes that should be of great value in any future fission product plant. How-

ever, the construction of future plants of this type will involve a very considerable cost. This cost will be balanced against a justifiable market demand, or against a sufficient improvement in the safety of waste storage at a particular site to justify the new practice.

In either case, the justification will be very sensitive to the scale of the reprocessing plant, that is, one will have to be generating very large quantities of waste before utilization or reduction to sealed source form can be economically justified over some much simpler method of waste handling.

Table 1 lists also the approximate extraction efficiency necessary for each specific nuclide in a typical waste after 10 years of storage before it is permissible to discharge that typical waste into an average environment. These efficiencies reflect both the relative concentrations of the nuclides and their individual biological hazard.

This is similar to the argument presented before you recently by Mr. Tomlinson.

If one were interested in recovering some of these fission products for utilization purposes, a 99 percent recovery would still leave behind sufficient activity so that the wastes are essentially just as hazardous as they were before recovery was attempted. Particularly for the case of strontium 90, recovery in the order of 99.999999 percent would be necessary. This is equivalent to Mr. Tomlinson's decontamination factor of 10 to the eighth. Also, in most cases the transuranics, americium 241 and plutonium 239, would be present in sufficient concentrations to require continued storage of waste. Thus it appears to me that even if fission product utilization were to be extensively practiced, unless trace amounts of noxious materials are also removed to an extremely high degree, utilization in itself could not be a solution to the waste disposal problem. Fission product recovery will have a beneficial effect on the waste disposal problem in this sense. By removing a large fraction of the fission products one removes the major source of heat generation in the storage tanks. Thus if fission product recovery is practiced, even though the residue from the recovery process probably cannot be discharged to the environment, the conditions for storing the residue become considerably less stringent than those required for storing the original high level waste. It should be pointed out, however, that even if one were to package and make use of some of the hazardous long-lived materials like cesium 137 and strontium 90, one would still face the problem of disposing of the radiation sources after they have become obsolete as a radiation device.

What, then, is a reasonable approach to the problem, and what part will fission product utilization play?

At the present time there are four sites in the United States where fuel is being reprocessed both for the military production program and for the power reactor program, i.e., Hanford, Savannah, Oak Ridge, and Idaho. In the foreseeable future it is unlikely that there will be more than, let us say, six such sites in the United States. In the past, through normal operational practice, once a single process has been put on a production basis, waste volumes have been reduced to a point where storage costs are a small fraction of plant operation cost. I suggest that, in time, when the power reactor program has stabilized and types of fuel elements have been standardized so that the fuel re-

covery process can be operated on a production basis, waste volumes from the power reactor program will also be reduced to the point where storage costs will be a small fraction of plant operation cost. If a major fraction of the fuel burned in this country is reprocessed at these six or more reprocessing plants, these sites will be the accumulators of a major fraction of the hazardous radionuclides generated.

The form of these wastes, from the point of view of minimum storage cost at a reprocessing site, is not likely to be the optimum form from the point of view of waste utilization. They may, for example, be in the form of a neutralized sludge, or may be absorbed on clay, et cetera.

I would not like at this time to propose what form these wastes should take, except to recommend that they be in a form considered safe for the particular conditions at that site by responsible individuals at that site and by responsible individuals in a regulating agency, for at least a single generation, and that they be in such form that each succeeding generation can make and carry out its own decision as to their disposition. For example, wastes can be stored cheaply and safely as neutralized sludges in monitored steel tanks for 20 years. Yet, at the end of that time, if some clever engineer has come up with some safer or more desirable and yet economic way of handling wastes, they can be transferred out of those tanks and converted to the new form.

One still faces the problem of wastes generated at reactor sites with attached reprocessing plants such as some homogeneous schemes or some pyroprocessing schemes. For these, consideration should be given during site selection to the potential accumulation of long-lived nuclides. I would recommend also that consideration be given to the development of techniques permitting the safe transportation of long-lived hazardous wastes either to the six or so aqueous reprocessing sites, or to some other limited number of national repositories.

Representative HOLIFIELD. In other words, these sites should not be chosen carelessly; they should have this very important point in mind?

Mr. MANOWITZ. That is correct. The difference I am trying to point out here is between reactors which make use of solid fuel elements which themselves can be shipped to a processing site, and the reactors which make use of some scheme whereby the reprocessing of cores is at that site itself.

These latter plants, then, will accumulate fission product rather than sending some fission product to another site where they will be accumulated.

So site selection is more important for these sites if they do intend to accumulate fission products, than for the solid fueled reactor sites.

Representative HOLIFIELD. They accumulate them. They have to take them out of the slurry in order to keep the homogenous type of plant running?

Mr. MANOWITZ. Yes, they continuously remove them, but if they have an attached reprocessing plant then either they will store them there or transport them some place else.

They transport them in this case in a form which is not as contained as it would be if you had them in a fuel element.

As time goes on, it is very likely that at these six or so major reprocessing sites, plants and equipment will become obsolete or not needed for the major separation job, and thus will become available for fission product recovery. It is my understanding that this is the situation at Hanford today. Under these conditions it may be possible to recover fission products on a very appreciable scale without too much of a first cost. This practice may then be justified on the basis of further developing the technology of fission product recovery, providing material for the development of fission product appliances, and on the basis of partially alleviating the waste disposal problem at that particular site.

As far as fission product utilization is concerned, the uses of cesium 137 and strontium 90 which tend to disperse them should be so controlled that these materials are in an extremely stable form, and in extremely safe containment. Uses requiring large quantities of strontium 90 and cesium 137 should be under particularly careful control and security, and should preferably be relegated to remote locations. On the other hand, under carefully controlled conditions, I would encourage the widespread use of the relatively safe and abundant radio nuclides like prometheum 147, krypton 85, ruthenium 106, cerium 144, zirconium 95, barium 140, and iodine 131. In this way advantage can be taken of our potential radiation resources, at the same time minimizing the danger to our future generations.

Mr. Chairman, I would like also to make some statement on the urgency of the waste disposal problem, reflecting some of the comments I have heard here from your committee and from previous witnesses.

Qualified people can have different opinions in this area but it is my personal opinion that there is no immediate urgency to the waste disposal problem. It has been mentioned that there is a considerable amount of gallonage available for waste at existing sites.

It must be remembered that a large fraction of the contribution to those waste tanks is from the weapons production program and that the power reactor program will contribute in my opinion, a relatively small fraction of waste to those volumes.

It is my feeling that what is needed is more emphasis on a widely divergent research and development program so that we can get the ideas of anyone who has good ideas in this field rather than emphasize a crash program of any sort in any particular system.

Representative HOLIFIELD. In other words, you think the facilities that we have in being at this time are going to be sufficient to take care of the waste material?

Mr. MANOWITZ. For some reasonable period of time. My understanding is that the power reactor fuels from these 17 or 20 reactors we are going to construct in the relatively near future will probably be handled at existing reprocessing sites, in fact, even the Euratom fuels will be handled at existing reprocessing sites.

Representative HOLIFIELD. That is true.

Mr. MANOWITZ. I don't think there will be any immediate problem as far as waste storage is concerned. I don't think even the Euratom systems will be any immediate problem.

So that in terms of time I think no crash program is needed but we do need a lot more information on new techniques for ultimate

disposal of these wastes and new techniques for interim disposal of these wastes.

Representative HOLIFIELD. I agree with you that we have enough tankage at this time, probably, for the current production for possibly a period of 10 years. But this again will depend on the acceleration of the reactor program.

Of course, it takes 3 or 4 years to build a reactor. If we go into an accelerated program of reactor building, why it might cut that 10 years down to 8 years or something like that.

This is certainly not too soon to start research and development.

Mr. MANOWITZ. No, sir; not at all.

Representative HOLIFIELD. I would agree with you what we need at this time rather than the building of extra plants which we will not need for 6 or 8 years or 10, we should get on with the research and development program and coordinate it and find out where we are going.

On the other hand, if we can develop within less than that time a safe way of storing these wastes underground, in salt mines, abandoned wells, or whatever it might be, why we can probably save a lot of money in the attempt to separate this material.

Mr. MANOWITZ. Yes, sir, I could not agree with you more on that either.

Chairman DURHAM. That is one of the main points. Of course, we started out with very little knowledge on this subject and we did the best thing we could, but we want to get along and get a less expensive program.

I agree with you that research and development is the only way you can arrive at it.

Mr. MANOWITZ. That was a bit of diversion from the fission product utilization field. I would be happy to answer any questions you have in that area.

Chairman DURHAM. You have made a very fine statement although a very technical statement.

Representative HOLIFIELD. You look with a dim view upon the value of the recovery as against the cost of recovery of usable isotopes?

Mr. MANOWITZ. No, sir.

Representative HOLIFIELD. You do not?

Mr. MANOWITZ. No, sir, I think some of these fission products will be recovered and on their own basis will be useful as a source of energy or source of radiation, but I don't think that is going to have very much of an effect on waste disposal.

Representative HOLIFIELD. That is true. You see no shortage of material either, in anticipation of the amount of recoverable, usable materials that might be in it; we certainly have no reason to think we are going to have a shortage and, therefore, we must preserve every gallon of this in the hopes that we will be able to use all the cesium and strontium that we might get out of it for peaceful purposes?

Mr. MANOWITZ. No. I think once again, that fission products particularly, are going to face competition with other ways of doing the job that one is now considering for the use of these particular materials.

Representative HOLIFIELD. There is no use in considering this a tremendously valuable bank of usable materials for the future, is there?

Mr. MANOWITZ. It all depends on the emphasis you put on tremendously valuable. I think this is a valuable bank that we should retain and for many reasons I think we should put these wastes in such places that we can get them out again when we want them, not only from the recovery point of view, but also from the long-term potential hazard point of view. These things, I think, go hand in hand.

Representative HOLIFIELD. They certainly go hand in hand if you put them in a 20-year tank. If you are putting them in a salt cave underground that might last for a thousand years, why then that point is not so important, is it?

Mr. MANOWITZ. That is right. My own opinion then would be, I see no need for putting them in a salt cave in the immediate future.

Representative HOLIFIELD. Of course, if you put them in a salt cave you could probably recover it.

Mr. MANOWITZ. If you could, then I withdraw my objection.

Representative HOLIFIELD. If you store it in solids you could and even if stored in fluids it could be pumped out, could it not?

Mr. MANOWITZ. I am interested only in the principle.

Chairman DURHAM. You do not object to research on the salt mines?

Mr. MANOWITZ. No, sir. I think all of these avenues should be investigated now from a research and development point of view and as many ideas as we have people that have ideas should be investigated.

Chairman DURHAM. You also emphasize the fact that the market development of these different products will probably face extensive competition from other things which can do the job just as well.

Mr. MANOWITZ. This is a hard world we are living in, yes, sir.

Chairman DURHAM. The reason I ask that question is that I read an article yesterday or the day before in some magazine where the headline says everybody is going to get rich off atomic energy. I thought that was misleading.

Representative BATES. Do you know when that plant at Stockton will become operational?

Mr. MANOWITZ. My understanding is sometime in 1960 is when the plant is to begin. I don't know whether it will actually become operational in that time.

Representative BATES. What is the future of that type of thing as far as you can foresee now?

Mr. MANOWITZ. I can only give you my own opinion in that area based only on the reading I have done in that area.

I think that in my own opinion once again, for the particular purpose that the Quartermaster Corps has, I am hopeful they are going to find very effective uses for radiation source.

Representative BATES. That is a fact today, is it not? It is just a question of degree.

Mr. MANOWITZ. I don't think it is a fact today. It has yet to be demonstrated on an appreciable scale.

Representative BATES. There are other factors like taste?

Mr. MANOWITZ. All of these things have yet to be investigated and overcome.

Representative HOLIFIELD. As I understand it, that experiment has been minimized considerably. I understand they are not going ahead, that they have cut down considerably on that.

Mr. MANOWITZ. My understanding is that they originally were thinking in terms of a reactor for doing this job. That has now changed and they are going to put in a radiation source made of cobalt 60 for doing it, the so-called high intensity food irradiator.

Chairman DURHAM. They went so far as to ask us for it and we authorized it.

Mr. MANOWITZ. Yes. My understanding is that construction is starting or will soon start. I think this is a good opportunity for comparing radiation source with high voltage machines since they are also going to put up a high voltage machine at the same site.

Representative BATES. Does that switch have any effect on speeding this up at all?

Mr. MANOWITZ. Yes, I think this particular operation will have an effect. It is the first opportunity for comparing production devices. Both the cobalt 60 device and the machine in this instance will be production devices. It will be the first opportunity for other people to realistically evaluate whether or not they can use devices of this sort for many types of radiation processes.

Representative BATES. Do they expect this will only be used for major purposes or do they expect to resolve these into smaller units for smaller purposes?

In other words, right now it would be a relatively large operation for the Army. Do they ever expect it to be used for, say, a fishing vessel? Do you know whether or not their thinking has advanced that far?

Mr. MANOWITZ. I would not be able to answer that question.

Representatives BATES. It would have to be made in a small package.

Mr. MANOWITZ. Yes. Depending on the specific application. I would not rule out small application as well as the large ones. It depends on the use you have in mind.

Representative BATES. I said fishing vessels.

Mr. MANOWITZ. You mean to sterilize fish, preserve them on the way to the market?

Representative BATES. Yes.

Mr. MANOWITZ. I can say it is possible. Whether or not it is practical, I don't know.

Representative HOLIFIELD. Thank you, sir.

Mr. DOAN. Mr. Chairman, could I make a statement along this line?

Representative HOLIFIELD. Certainly, Mr. Doan.

Mr. DOAN. First, with regard to the amount of available tank storage underground that we now have. This could be anything we want it to be just by putting in more tanks, so this places no limitation on anything.

Next, with regard to the urgency of finding better methods of disposal, I would call attention to the fact that most of the waste solutions in storage now are much less corrosive than the processing solutions for the zirconium fuels that many of the power reactors are going to at the present time. The life of the tanks used to store zirconium wastes is going to be much shorter, so that we can't live as comfortably

with long-time storage of zirconium solution wastes. We have to use better tanks, more costly tanks.

Third, with respect to the utilization of the fission products in the waste solutions. There is a wide range of opinion on what we are going to be able to do with them. I would call attention to the fact that what we can now see in the way of practical applications of byproduct radioisotopes is only a small fraction of what we will be able to see later. To draw a parallel with the oil industry, you will recall that not many years ago the gas produced along with the oil was a nuisance all over Texas and Oklahoma. One could see the gas flares burning the gas that the producers wanted to get rid of, so they could produce some oil. Today we have developed out of that same gas, which every one wanted to get rid of, a gigantic liquefied petroleum industry.

We have turned gas into plastics and fertilizers; and many of the automobiles that roll down the highways today do so on synthetic rubber tires made from the same gas that was a nuisance only a few short years ago.

Representative HOLIFIELD. Thank you, Mr. Doan.

I gather from your remarks that you are in the hope that there will be beneficial uses discovered in the next few years out of this material which we do not see at the present time.

Mr. DOAN. Yes, I think there is a good likelihood of that.

Representative HOLIFIELD. Our next and last witness will be Mr. A. F. Rupp, for ORNL.

Mr. Rupp, we are glad to hear from you.

STATEMENT OF A. F. RUPP,[9] THE OAK RIDGE NATIONAL LABORATORY, FISSION PRODUCT PILOT PLANT

Mr. RUPP. My talk is about a specific pilot plant that we built, rather than on this general subject.

The radioactive wastes generated by the operation of nuclear reactors are largely fission products which might more properly be called byproducts, since they are potentially valuable materials. The fission-product mixture contains many radioisotopes that have already been found to be extremely useful. The use of radioisotopes as tracers in research, for radiography and in medical therapy, are well known.

While most of the work with radioisotopes up to the present time has been done with relatively small quantities of material (hundreds of curies), experiments on the pasteurization of foods, retardation of sprouting in vegetables and grain, sterilization of medical supplies, promoting of chemical reactions, and special heat-power sources indi-

[9] Date of birth: Nov. 4, 1911. Place of birth: Lawrenceburg, Ind. Education: Graduated from Purdue University, 1933, B.S. Ch. E. Work history: 1933–40, J. E. Seagram & Sons, general chemical engineering. 1941–45, DuPont Co., research and development. Explosives, Manhattan project, Clinton Laboratories, and Hanford. 1946 to date, Oak Ridge National Laboratory, Union Carbide Nuclear Co., supervising design, development, and operation of high-level radiochemical processes; reactor operations; engineering and mechanical work, and inspection engineering. Present capacity: Superintendent, laboratory services. Publications: Contributing author to "Radioisotopes in Industry" (Rheinhold). Contributing author to "Progress in Nuclear Energy," series III, vol. I. "The Preparation of Radioisotopes." Contributing author to "Nuclear Engineering Handbook" (Etherington). "Production of Radioisotopes." Author of various papers published on the production of radiosotopes, handling radioactivity on large-scale separations of fission products, and economics of byproduct sources. Member in societies: American Chemical Society, American Institute of Chemical Engineers.

cate that millions of curies of radioisotopes may be needed in the future. Market surveys are now being conducted by several commercial organizations to determine the probable market for fission products in the fairly near future. We do not as yet have complete results on these surveys. In order to test proposed processes, it was considered necessary to have reasonably large quantities of radioactive material, that is, thousands of curies.

In other words, one of the reasons we wanted to go into pilot plant operations is that if you want to find out whether something is good for anything, you have to have something to test first; our motivation was different than disposing of waste. We were in the business of trying to make some useful material to check.

About 10 years ago, work on a very small scale was started at ORNL to investigate methods for removing the important long-lived fission products from waste solutions produced in processing spent reactor fuel elements. At that time Cs^{137}, which has a 30-year half life and emits penetrating gamma radiation, was considered the most valuable fission product, although work was also done on the removal of 28-year Sr^{90}, 290-day Ce^{144}, 2.6-year Pm^{147}, 1-year Ru^{106}, and 200,000-year Tc^{99}. In this case I am referring of course to the half life of the material. Because the original work was directed toward medical uses, it was decided to separate the fission products in as nearly pure form as possible. This decision has since proved valuable, as it also appears desirable to prepare industrial radiation sources in fairly pure form. The advantages of purified material are a reduction in loss of radiation energy by self-absorption, accurate knowledge of decay rates and radiation characteristics, and avoidance of mixing fission products of greatly differing toxicity (e.g., Cs^{137} and Sr^{90}).

After the recovery processes were taken through all stages of laboratory and semiworks development, construction of a full-scale pilot plant was started in 1955, and this small plant is now in first phases of operation (fig. 1). The nominal plant design is for an annual output of 200,000 curies of Cs^{137}; other fission products will be separated in varying amounts, depending upon the need for the products, the direction taken by the research program, and the characteristics of the waste feed material.

The main objective of the fission product pilot plant (known as F3P) is to recover the six fission products previously mentioned and to fabricate useful kilocurie sources from the purified fission product compounds. For example, Cs^{137} sources are made by compressing pure, dry cesium chloride powder into pellets (fig. 2) and hermetically sealing them in stainless steel containers. Incidentally, we do that as a double encapulation. We made a source 3½ years ago of this kind, which has been in a medical unit at ORNL and there has been no difficulty whatsoever with it—no leakage of any kind.

I brought along some material to give you an idea what this stuff looks like. Of course this is a stand in, it is not radioactive but it is the actual chemical material. This is what a thousand curies looks like. This is what it is like when pressed.

Representative PRICE. Mr. Rupp, I don't want to interrupt your statement, but during the first trip I ever made through Argonne back

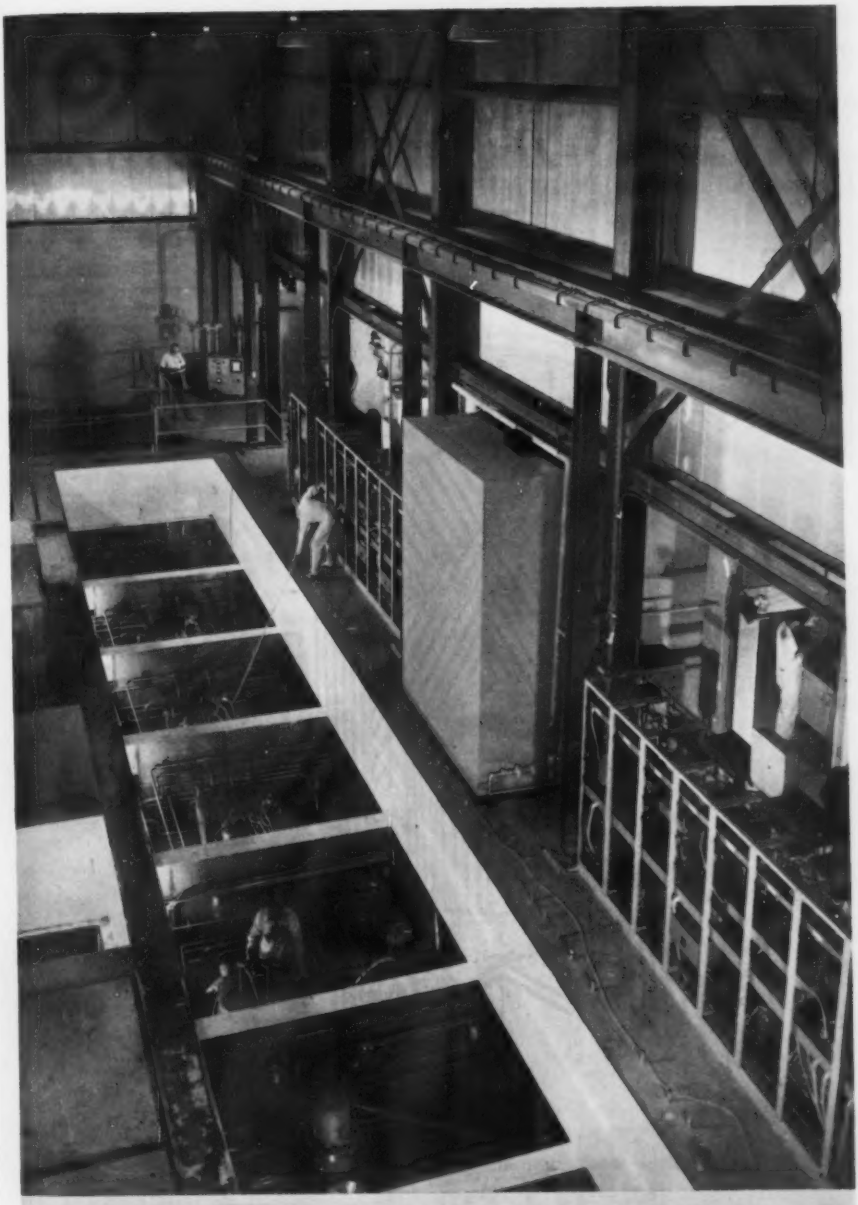

Figure 1. View of chemical cells in fission product pilot plant.

in 1946 they had a project known as Redox. Is that an additional phase in experiments in recovery?

Mr. RUPP. No, that was a process developed for processing spent uranium fuel containing aluminum—a special process for recovering uranium from fuel elements. It does produce waste, though. It produces a waste with aluminum in it that would have to be processed. It is not the same process.

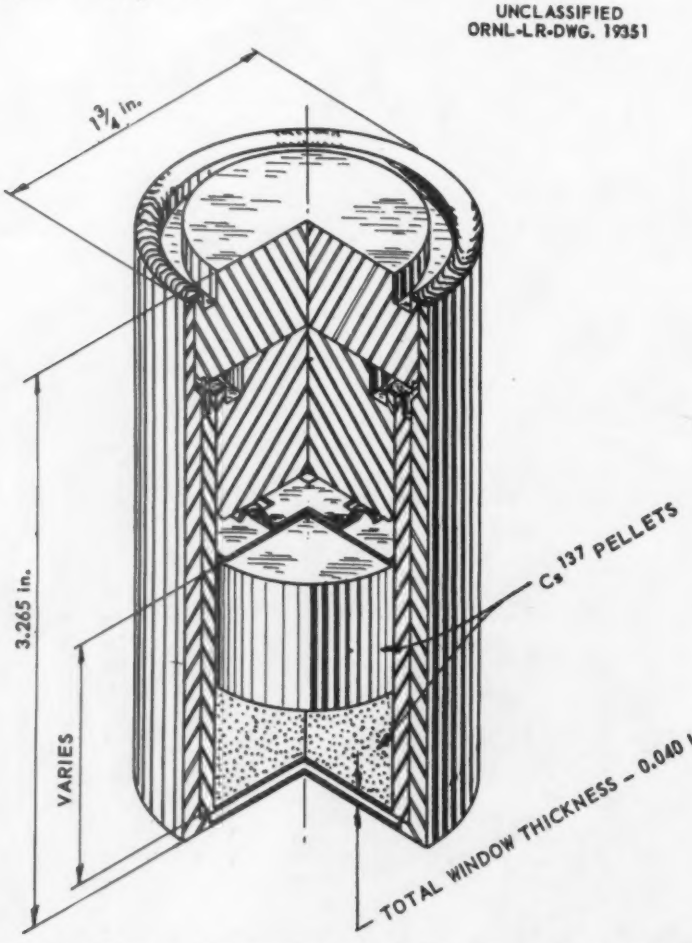

FIGURE 2.—Stainless steel encapsulated Cs 137—Source for medical therapy.

The main chemical separations are made by several series of precipitations performed on the highly concentrated liquid waste. The gross precipitation steps alone achieve a concentration factor greater than 10 in the radioactive components of the waste (see fig. 3). By using reactions which have high individual yields and by feeding back the small amounts removed in the tail end of the process to the main processing stream, good product recovery and decontamination of the waste stream are achieved.

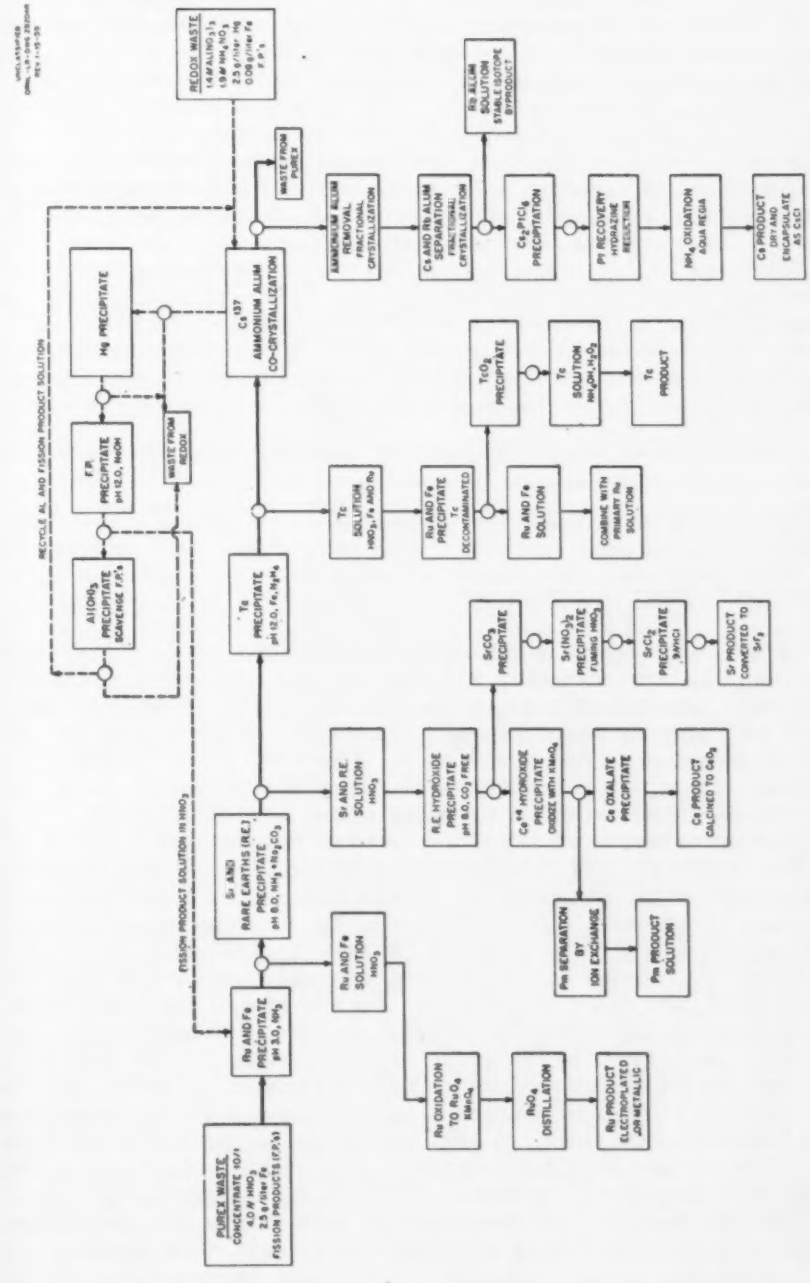
PUREX WASTE
CONCENTRATE 10:1
4.0 M HNO3
2.5 g/liter Fe
FISSION PRODUCTS (F.P.'s)
Ru AND Fe PRECIPITATE
pH 3.0, NH4OH
FISSION PRODUCT SOLUTION IN HNO3
Sr AND RARE EARTHS (R.E.) PRECIPITATE
pH 9.0, NH4OH+(NH4)2CO3
Ru AND Fe SOLUTION HNO3
Ru OXIDATION TO RuO4 WITH NaOCl
RuO4 DISTILLATION
Ru PRODUCT ELECTROPLATED OR METALLIC
Sr AND R.E. SOLUTION HNO3
R.E. HYDROXIDE PRECIPITATE pH 8.0, CO3 FREE
Ce+4 HYDROXIDE PRECIPITATE OXIDIZE WITH NaOCl
SrCO3 PRECIPITATE
Sr(NO3)2 PRECIPITATE FUMING HNO3
SrCl2 PRECIPITATE 9 M HCl
Sr PRODUCT CONVERTED TO SrF2
Pm SEPARATION BY ION EXCHANGE
Pm PRODUCT SOLUTION
Ce OXALATE PRECIPITATE
Ce PRODUCT CALCINED TO CeO2
Tc PRECIPITATE pH 12.0, Fe, Ni2Fe4
Al(OH)3 PRECIPITATE SCAVENGE F.P.'s
F.P. PRECIPITATE pH 12.0, NaOH
Hg PRECIPITATE
RECYCLE AL AND FISSION PRODUCT SOLUTION
WASTE FROM REDOX
Cs137 AMMONIUM ALUM CO-CRYSTALLIZATION
WASTE FROM PUREX
REDOX WASTE
14 F Al(NO3)3
1.9 M NH4NO3
2.5 g/liter Na
0.06 g/liter Fe
F.P.'s
AMMONIUM ALUM REMOVAL FRACTIONAL CRYSTALLIZATION
Cs AND Rb ALUM SEPARATION FRACTIONAL CRYSTALLIZATION
Rb ALUM SOLUTION STABLE ISOTOPE BYPRODUCT
Cs2PtCl6 PRECIPITATION
Pt RECOVERY HYDRAZINE REDUCTION
NH4 OXIDATION AQUA REGIA
Cs PRODUCT DRY AND ENCAPSULATE AS CsCl
Tc SOLUTION HNO3, Fe AND Ru
Ru AND Fe PRECIPITATE Tc DECONTAMINATED
TcO2 PRECIPITATE
Tc SOLUTION NH4OH, H2O2
Tc PRODUCT
Ru AND Fe SOLUTION
COMBINE WITH PRIMARY Ru SOLUTION

FIGURE 3.—Separation of fission products from reactor waste.

I might mention here that we have worked chiefly on a waste called Purex waste which contains only the fission products, and nitric acid, which is volatile, and corrosion products; so we can evaporate it down to a very small volume before we start—the economies that one can make in such a process are tied up in steps of that kind.

Representative DURHAM. The evaporator contents remain stable?

Mr. RUPP. You can take it down as much as 20 to 1 and still work with it. We are going to try to see how far down we can take it and still work with it. The main trouble is that we get too much heat produced. For example, in a centrifuge you get steam produced which blows the material off the wall and things of that kind. So we don't know what the limitations are.

While the F3P is primarily intended to demonstrate the recovery of relatively large quantities of individual fission products, it at the same time decontaminates the waste stream fed into the plant. This is illustrated in the following table, which gives rough estimates on the removal of the main radioactive components of the waste stream when the plant is operated to achieve maximum decontamination:

Fission product plant decontamination of waste (using Purex waste)

Fission product	Cs^{137}	Sr^{90}	Rare earths	Ru
(Decontamination factor (D.F.) ordinary recovery operations).	10^4	30	10^2	10^2
Extra precipitation steps, D.F		10^3	10^4	10^2
Ion exchange cleanup, D.F	10^6	10^4	10^4	10^3
Total D.F. possible	10^{10}	3×10^7	10^{10}	10^7

I point out we are working here only with Purex waste. Ordinarily we would be concerned only with the recovery operations and would not try to put in the cleanup steps, but one could, and we have taken most of these processes through these cleanup steps and can get decontamination factors of this kind. The other speakers today have contended that this is uneconomical. We contend that in the case of one waste, Purex waste, it probably is not uneconomical.

The figures here mean, if you use the total decontamination factor on cesium or strontium, if 10 billion curies are put through such processes, only 1 curie would come out in the waste streams. If the refined products are compressed and doubly sealed by welding into convenient corrosion-resistant metal packages, the material can be used in irradiation devices or stored in isolated underground vaults where it can be kept under permanent surveillance. Should any of the packages develop leaks, as determined by radiation detection devices, the offending package can be removed for repair. In other words, we are advocating not putting all the eggs in one basket. We advocate putting them in packages, in individual containers in underground vaults, where you can keep an eye on them. While this method of concentrating to the minimum volume is of value to the waste disposal program, it is also expected that uses will be found eventually for a large portion of the radioactive material. If uses develop only slowly, the radioactive material is in safe storage and available for use when needed.

It is instructive to note that the concentrated fission products can be stored in a relatively small volume. For example, the total weight of all the fission product elements resulting from the operation of a

1000-megawatt (heat) plant for 1 year is about 750 pounds. If all these elements were separated and converted into chemical compounds (chlorides, oxides, or fluorides), then dried and compressed to a density of 3 g/cc, they would occupy a volume of about 5 cubic feet. The compounds of the two important long-lived isotopes, Cs^{137} and Sr^{90} would occupy less than 1 cubic foot when compressed to the maximum practical density. Since the total volume of waste from which the fission products were removed would be about 540,000 gallons, or 73,000 cubic feet (natural uranium process waste), this represents a concentration of the volume in which the most toxic long-lived activities are contained by a factor of about 10^4. The quantities per year from the above 1000-megawatt plant are listed below:

Element	Weight (pounds)	Radioisotope	Curies (after 1 year)	Compressed volume (cubic feet)
Cs	87.6	Cs^{137}	1.27×10^6	0.6 (CsCl).
Sr	30.0	Sr^{90}	1.12×10^6	0.2 (SrF_2).
Ce	91.0	Ce^{144}	21.3×10^6	0.25 (CeO_2).
Ru	40.6	Ru^{106}	1.1×10^6	0.05 (Ru Metal).
Pm	13.7	Pm^{147}	4.6×10^6	0.05 (Pm_2O_3).
Tc	21.4	Tc^{99}	166	

I brought along also some samples of the other materials just to give you an idea about what they look like. This is almost 2,000 curies of cerium that is put in its final form (CeO_2) sintered.

As is the case with all radioisotope production at ORNL, the F3P is expected to produce refined fission products on a full cost-recovery basis, including depreciation, overhead, and allowance for other normal business expenses, but allowing no credits for disposing of the waste processed. The prices recently set for fission products from F3P when purchased in fairly large quantities are as follows:

	Price per curie
Cesium 137	$1.00
Promethium 147	1.75
Cerium 144	1.00
Strontium 90	5.00
Technetium 99	1,360/gram

Source encapsulation and packaging charges are extra. While these prices are a factor of 10 to 100 lower than previous ones, it is expected that further reductions can be made when production increases beyond the pilot plant stage. If such a plant were operated mainly for waste disposal purposes, the fission products could be separated individually or in groups in fairly impure form, thus allowing a considerable reduction in cost. If the total amount of radioactive products from processing wastes from the 1,000-Mw reactor were sold for an average of only 10 cents per curie, the gross income would be several million dollars per year—more than enough to pay the processing plant expenses. While we cannot predict that a market will develop for all the radioactive material produced from a large nuclear industry, it appears that a portion of it will be needed; and the sale of these products may produce enough extra income to store the rest, particularly Sr^{90}.

Representative HOLIFIELD. Thank you.

Mr. RUPP. If you would care to look at these samples, I will pass them around.

Representative HOLIFIELD. Are there any questions of Mr. Rupp?

Representative DURHAM. What you are actually doing in this experimental program is to reduce the volume.

Mr. RUPP. We are taking it down as far as you can go; and, in several cases, for certain of uses we really do have to have it in this pure form.

Representative DURHAM. How much did you start out with in volume to begin with on that?

Mr. RUPP. That represents probably several thousand curies of strontium. It was several thousand gallons.

Representative DURHAM. What is the difference in curies? Can you estimate it in the volume you started with and what you wound up with?

Mr. RUPP. In this particular case if we had taken it all through the process I mentioned with the ultimate waste disposal steps included, we would come out with roughly something like a millionth of the activity per unit volume that we started with, that is, in waste.

Representative DURHAM. Then you lost curies on the way down.

Mr. RUPP. No, a millionth of the original would be in the waste. The rest of it would be in the product somewhere.

I should like to point out that there are many wastes. For example, Dr. Doan mentioned some very formidable wastes coming up that are very difficult to process. We have been concentrating on Purex waste and it would be good if all the wastes were that pure to begin with. It would certainly simplify the problem.

Representative DURHAM. When you speak of Purex you are of course speaking of the nitric acid process.

Mr. RUPP. That is right. It is a very good waste to work with from the standpoint of production of fission products.

Representative HOLIFIELD. You picked the easiest one.

Mr. RUPP. Yes, we picked the easiest one. It is a very tough problem so we picked the easiest one to start with.

Representative DURHAM. Why did you pick nitric acid?

Mr. RUPP. We didn't pick that. The person who designed the uranium process for removing uranium from spent fuel picked that for what they call a salting agent instead of aluminum nitrate.

Representative DURHAM. I was thinking of the effect of nitric acid on organic matter.

Mr. RUPP. They don't have trouble of that kind. It is not that concentrated.

Representative DURHAM. Sulfuric acid?

Mr. RUPP. Sulfuric acid they don't use much at all. There is only one process it has been considered in.

Representative DURHAM. It is the father of all acids that have been spoken of.

Mr. RUPP. Indeed it is.

Representative DURHAM. What is that sample?

Mr. RUPP. That is ruthenium plated on copper. I put that in to show you what could be done. That is electroplated. Ruthenium can be electroplated on a large surface that can be used as a beta source

for sterilizing bandages, for example, things of that kind. We intend to make up sources of that kind.

Representative DURHAM. I suppose it is about as valuable as gold.

Mr. RUPP. It is a seminoble metal. We are going to recover and store the ruthenium. In about 10 years it will all be decayed and we will take the palladium out of it. Palladium is valuable. There are several elements I don't have exhibited because they don't exist on earth in forms other than the radioactive ones. These are promethium and technetium. The only way I could have brought some of those would have been to have brought the real thing because they don't exist on earth in a stable form.

Representative HOLIFIELD. Are there any further questions of Mr. Rupp?

If not, we will resume our hearing in the old Supreme Court room, room P–63, on Monday, February 2 at 10 a.m. We will have a series of witnesses on this subject of future estimates and economics of waste disposal, and summary and evaluation of environmental factors, industrial viewpoint and a roundtable discussion. On the 5th day we will continue and that should be the last day if we are able to get to the end of the witness list, otherwise we will have to carry over.

Before we adjourn I have a paper which I will place in the record here.

(The material referred to follows:)

FISSION BYPRODUCTS UTILIZATION—OFFICE OF ISOTOPES DEVELOPMENT, U.S. ATOMIC ENERGY COMMISSION

Objective of the Commission's isotope development program is to bring about widespread commercial scale use of radioisotopes and high-intensity radiation. Development of markets for multimegacurie amounts of fission byproducts and other high intensity radiation sources will be beneficial to the U.S. economy as a whole. It will not significantly alleviate the technical problems or requirements for ultimate waste disposal. At best, fission byproduct utilization wil defray only a very small fraction of waste storage costs directly. This is because (*a*) the multimegacurie quantities of fission byproducts required for commercial scale uses would still represent only a fractional percentage of total reactor effluents; (*b*) 100 percent removal of long-lived fission byproducts desired for industrial use, necessary to permit eventual environmental disposal of the shorter-lived constituents of reactor effluents, would be prohibitively expensive, and (*c*) industrially employed, long-lived fission byproducts ultimately will have to be returned to disposal channels after they have outlived their usefulness. Indirectly, however, increased national income resulting from fission byproduct utilization may more than cover the costs of waste storage and ultimate disposal.

The program for high-intensity radiation development is twofold: (*a*) research and development work that will lead to large markets for megacurie sources, and (*b*) research and development work that will stimulate commerical interest in supplying radiation sources (whether by chemical processing of fission byproduct wastes or through other techniques).

Utilization development activities include investigation of (*a*) theoretical aspects of radiation interaction with matter as they pertain directly to the gainful uses of radiation energy, (*b*) production and fabrication of radiation sources (radiation source technology) and design of radiation facilities (radiation engineering), and (*c*) specific radiation products and processes having commercial potential.

Production development activities include investigation of (*a*) technology of fission byproduct separation, (*b*) market potentials for various fission byproducts, and (*c*) problems directly related to integrating radiation processes into the national industrial complex.

These utilization and production development activities are being conducted by the AEC Office of Isotopes Development, at AEC National Laboratories, and through contract with private organizations, including the CEM group (Emerson Radio & Phonograph Corp., General Aniline & Film Corp., and Revere Copper & Brass, Inc.), Arthur D. Little, Inc., Radiation Applications, Inc., Battelle Memorial Institute, and William H. Johnston Laboratories, Inc.

Success in the foregoing program should establish radiation processing in the chemical, plastics, pharmaceutical, food, and other industries on a sound technological and economic footing.

(Whereupon, at 4:15 p.m. Friday, January 30, 1959, the hearing was recessed, to reconvene at 10 a.m. Monday, February 2, 1959, in room P–63, the Capitol.)

INDUSTRIAL RADIOACTIVE WASTE DISPOSAL

MONDAY, FEBRUARY 2, 1959

CONGRESS OF THE UNITED STATES,
SPECIAL SUBCOMMITTEE ON RADIATION,
JOINT COMMITTEE ON ATOMIC ENERGY,
Washington, D.C.

The special subcommittee of the Joint Committee on Atomic Energy met at 10 a.m., pursuant to recess, in room F–63, the Capitol, Hon. Chet Holifield, chairman of the special subcommittee, presiding.

Present: Representatives Holifield and Hosmer, Senators Anderson, Dworshak, and Aiken.

Also present: James T. Ramey, executive director, David R. Toll, staff counsel, and Dr. Walton Rodger, technical consultant, Joint Committee on Atomic Energy.

Representative HOLIFIELD. This morning the Special Subcommittee on Radiation is continuing these public hearings on industrial radioactive waste disposal. Last week we received statements on the PWR reactor. Then we received testimony from scientists carrying out the research and development program, both with respect to low- and intermediate-level waste, and high-level waste.

This morning we are scheduled to hear Dr. Bruce on future estimates and economics of waste disposal, and then from five leading authorities to summarize and evaluate the possible effects of future quantities of radioactive waste on man and his environment. If the time permits we hope to have a roundtable among the various experts to bring out points of agreement and disagreement, and exchange of ideas.

Our first witness this morning is Dr. Frank R. Bruce, of the Oak Ridge National Laboratory. We are glad to have you with us this morning, Dr. Bruce. Would you please come forward and proceed?

STATEMENT OF DR. FRANK R. BRUCE,[1] OAK RIDGE NATIONAL LABORATORY, OAK RIDGE, TENN.

Dr. BRUCE. Mr. Chairman, committee members, changing emphasis from the production of fissionable material for weapons purposes to the use of fissionable material for power generation is accompanied by a corresponding change in the nature and magnitude of the waste treatment and disposal problems anticipated. These problems stem,

[1] Born in Beverly, Mass., Feb. 9, 1919. He obtained the bachelor of science degree in chemistry from Tufts College in 1942. Since 1943, he has been associated with the Oak Ridge National Laboratory, and is associate director of the chemical technology division. His experience is primarily concerned with the research and development of separations processes for uranium, plutonium, and fission products. He is also an editor of "Process Chemistry," a volume of the "Progress in Nuclear Energy" series.

first, from the fact that power reactor fuels are designed for extreme corrosion resistance and must be dissolved in highly corrosive reagents prior to chemical processing; second, the rapid growth of the nuclear power industry will result in very large quantities of waste over the long term. Finally, it is desirable to carry out fuel reprocessing and, if possible, waste disposal operations near the centers of power consumption, which are expected also to be centers of high population density. The purpose of this paper is to assess some of the elements of waste treatment and disposal that are expected to become important in the development of the nuclear power industry of the future.

GROWTH OF THE NUCLEAR POWER ECONOMY

The magnitude of the waste disposal problem of the future depends on the size of the nuclear power economy. For the purpose of this

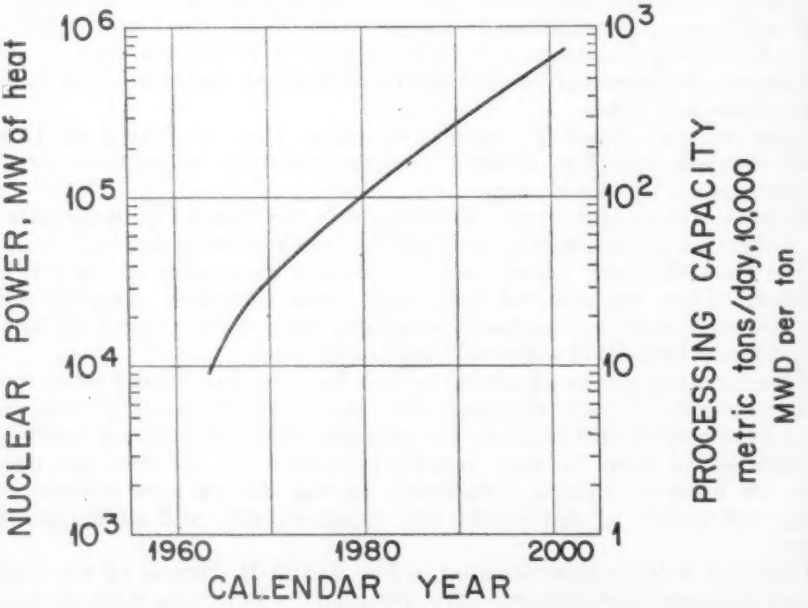

FIGURE 1.—Predicted power growth and fuel reprocessing capacity.

study the estimate of J. A. Lane, (1) shown in figure 1, is used primarily because it falls close to the average of all estimates. According to Lane, about 33,000 megawatts of nuclear heat will be installed by 1970 and nearly 300,000 megawatts by 1990. By the year 2000, about 700,000 megawatts will be installed. These figures established the quantities of fission products that will be produced and that must be stored.

It is expected that natural or slightly enriched power reactor fuels, on the average, will be irradiated to a level of about 10,000 megawatt days per ton of uranium, at which time they will be reprocessed for recovery of residual fissionable material. The quantity of fuel that must be reprocessed, also shown in figure 1, determines the volume of waste produced. In 1970, fuel will be processed at the rate of 3.3

tons per day, in 1990 at 30 tons per day, and by 2000 at 70 tons per day.

F. L. Culler has described the aqueous processing techniques in use for current fuels and has presented the available information on the newer techniques, including pyrometallurgical processing and fluoride volatility, which may become important in the future. In this discussion it is assumed that fuel dissolution by aqueous methods and solvent extraction of the solution will be used.

COMPOSITION AND QUANTITIES OF WASTE ANTICIPATED

The chemical composition and volume of waste produced by the nuclear power industry will be determined by the processing technique used, which, in turn, will be determined by the fuel composition. In this study it is assumed that the power reactors of the year 1980 will fall into the grouping shown in table 1. Ten percent of the total power is assumed to be produced by breeders and 90 percent by converters. Of the 10 percent produced by breeders, 5 percent is expected to be derived from fast breeders using stainless steel-clad fuel, and 5 percent from homogenous breeders. Of the 90 percent produced by converters, 40 and 25 percent are expected to come from reactors using zirconium-clad uranium fuels and stainless steel-clad uranium fuels, respectively 20 percent from thorium converters using zirconium-clad fuel and 5 percent from thorium converters using carbon-clad fuel.

TABLE 1.—*Assumed distribution of reactor types in 1980*

Reactor type	Fuel	Percent of nuclear power
Breeders:		
Fast	Stainless steel clad	5
Homogeneous	Aqueous	5
Converters:		
Uranium	Zirconium clad	40
	Stainless steel clad	25
Thorium	Zirconium clad	20
	Carbon clad	5
Total		100

COMPOSITION OF POWER REACTOR FUEL WASTES

Table 2 shows the important constituents in the anticipated wastes (3) from the processing techniques that will be used for the recovery of spent reactor fuel. These processes are under development but are ready to be demonstrated on an engineering scale. The techniques now used for the present aluminum-clad uranium and aluminum-uranium alloy fuels have already been described. The point I should like to make from this table is that in the case of the stainless steel- and zirconium-clad uranium dioxide fuels, the approach is to remove chemically the jacketing material, stainless steel, and zirconium. The core material, uranium dioxide, is then dissolved in nitric acid. From this type of natural uranium and slightly enriched uranium fuel we then have two types of waste: (1) a waste from the jacketing material; and (2) a waste from the core material. The radioactivity associated with the decladding solution is about 1,000 times less than the radioactivity associated with the high-level waste. However, there is still

sufficient radioactivity associated with the decladding solution that it must be handled as a high-level waste.

In the case of the zirconium-uranium alloy fuel, the entire fuel is dissolved together, since the two are intimately mixed. The same is true of the stainless steel-uranum dispersion fuels.

The wastes expected from power reactor fuels differ from current wastes mainly with respect to their content of dissolved stainless steel, zirconium, and fluoride, but they will be less amenable to tank storage than are the simpler wastes resulting from present-day processing of aluminum-clad uranium or uranium-aluminum alloy. Present waste disposal practice usually entails neutralization of waste with caustic prior to storage, but solutions of stainless steel, zirconium, and aluminum form voluminous precipitates on neutralization. The high heat generation rate anticipated in power reactor wastes and the presence of precipitates may require special provisions to remove heat and to prevent bumping if the wastes are stored in tanks. In addition, the reagents used to dissolve the power reactor fuels create serious corrosion problems in the long-term storage of the wastes.

TABLE 2.—*Composition of wastes anticipated from various types of reactors*

Fuel type	Decladding solution	High-activity waste
Natural U:		
Aluminum-clad U	$NaAlO_2$, 1.2 *M*	HNO_3, 2 *M*.
Stainless steel-clad UO_2	(Fe, Ni, Cr) SO_4, 80 g/liter	HNO_3, 2 *M*.
Zirconium-clad UO_2	Zr^{++}, 0.5 *M*; NH^+_4, 1.5 *M* F^-, 3.7 *M*; NO^-_3, 0.6 *M*.	HNO_3, 2 *M*.
Enriched U:		
Aluminum-uranium alloy		HNO_3, 1.3 *M* $Al(NO_3)_3$, 1.6 *M*
Zirconium-uranium alloy		HNO_3, 3.2 *M* Zr^{++}, 0.8 *M* NH_4F, 1.8 *M*
Stainless steel-uranium cermet		Stainless steel, 50 g/liter $Al(NO_3)_3$, 0.12 *M* HNO_3, 3 *M*

QUANTITIES OF HIGH-ACTIVITY WASTE EXPECTED

Table 3 shows the volume of waste anticipated from the various power reactor fuel types. The magnitude of the future waste disposal problem depends on the direction of fuel development, with the addition of alloying elements increasing the volumes of waste that must be stored, as well as on chemical processes used to recover the irradiated fuel. In the case of the natural or slightly enriched fuels, the volumes of waste to be stored are generally much lower than for the highly enriched fuels. This stems from the fact that in the former fuels, uranium 238, present as a diluent for the uranium 235, is extracted out of the waste stream in fuel reprocessing. In the case of the highly enriched fuels the diluent, a nonradioactive alloying material which is usually present, remains with the waste stream and contributes to the disposal problem.

Present indications are that the uranium oxide and mixed uranium-thorium oxide fuels will be widely used. This is the type shown here. This type yields roughly tenfold less waste volume per unit of heat extracted than an alloy fuel such as that used in the naval reactors.

TABLE 3.—*Volume of waste to be stored from power reactor fuels*

Fuel type	Waste volume, gallons	
	Per kilogram of uranium	Per megawatt-day
Natural or slightly enriched:		
Aluminum-clad uranium	[a] 0.33	0.033
Stainless steel-clad UO_2	[a] 1.06	0.106
Zirconium-clad UO_2	[a] 1.46	0.146
Highly enriched:		
Aluminum-uranium alloy	125	0.5
Zirconium-uranium alloy	300	1.2
Stainless steel-uranium cermet	65	0.26

[a] 0.06 gallon is high-activity waste; the balance is decladding.

Figure 2 shows the waste volumes that will be accumulated in the future. By the year 1980, 36 million gallons of waste will have been accumulated. By way of comparison, it has been reported that at the Hanford site about 50 million gallons of waste is stored in underground tanks. Thus, it appears that even by the year 1980 the wastes accumulated from the nuclear power industry will be much less than the volume now stored at Hanford.

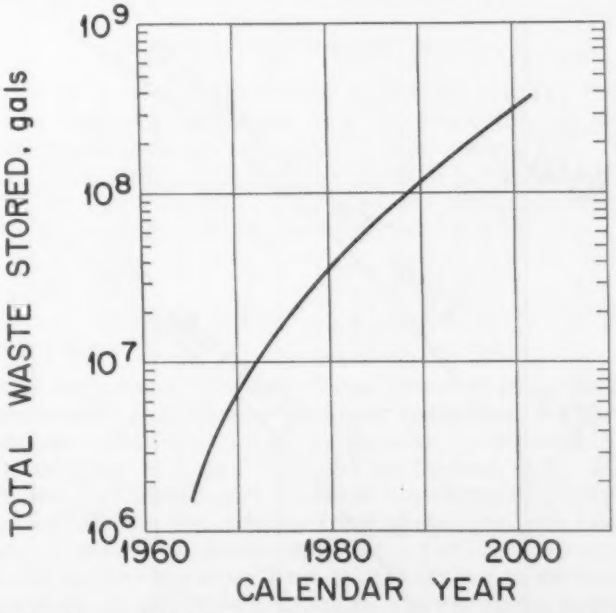

FIGURE 2.—Accumulated volume of high and intermediate level waste.

Prospects for markedly decreasing the volumes of waste resulting from power reactor fuels lie in two areas. First, mechanical processing methods are under development for separating cladding agents from fuel cores. Since chemical dissolution of cladding contributes the largest fraction of the volume to be stored, the mechanical techniques, if perfected, will result in a marked reduction in waste volume. Secondly, methods for converting power reactor wastes to a solid

form prior to storage are being studied. In the case of a zirconium-clad uranium dioxide fuel, which produces about 1,460 gallons of waste to be stored per ton of fuel, the stored volume could be reduced to 25 gallons per ton by conversion of the waste to a solid. Not only would this process reduce the volume of waste to be stored, but it would also eliminate the bumping and corrosion problems associated with the storage of the waste in liquid form. At the same time, storage of liquid waste introduces new problems of heat removal and evolution of radioactive gases.

The combination of the high toxicity of the fission products and their expected production in very large quantities in the nuclear power industry makes it essential that methods for their containment be absolutely safe. Figure 3 shows the accumulated radioactivity resulting from the predicted nuclear power industry (4). A feeling for the magnitude of the hazard potential associated with the waste may be obtained by considering the following. By the year 1980, 10 billion curies of activities will be stored; 800 million curies of strontium 90 will be contained in 36 million gallons of solution.

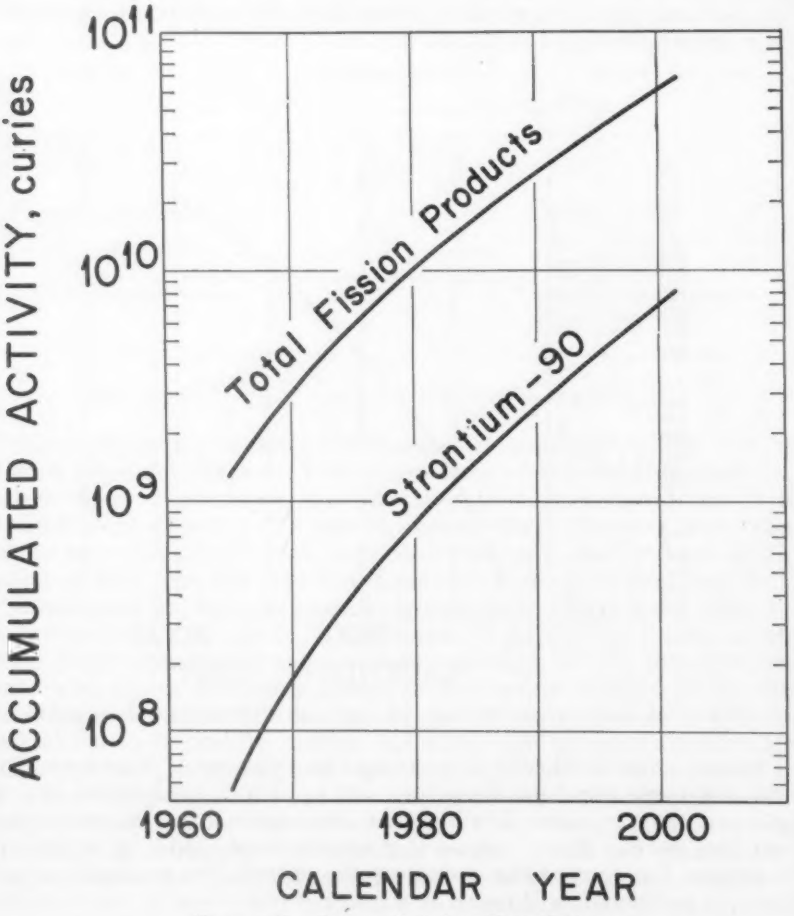

FIGURE 3.—Accumulated radioactivity.

GASEOUS WASTES

Fission product gases appear in the off-gas system from fuel reprocessing plants. The only important one from the standpoint of long-term waste disposal is krypton 85, which has a 10-year half life. Iodine 131 is by far the most significant when short-decayed fuel is processed. Although processes are currently in use to remove iodine from chemical plant off-gas systems, the present techniques probably do not achieve complete enough removal to permit locating chemical plants in congested areas. Further process development is required in this regard.

Although the hazard associated with krypton is slight, its continued buildup necessitates developing methods for removing it from power reactor fuel reprocessing off gases. By the year 2000, 700 megacuries of krypton 85 will have been released to the atmosphere (5). This quantity mixed with the air surrounding the earth to a height of 10 miles would increase the background by 4.2 percent assuming that the air background is now 0.015 milliroentgen per hour. While it appears that the release of rare gases to the atmosphere may be permissible during the early stages of the nuclear power industry, it will eventually be necessary to remove the rare gases from chemical plant waste and retain them for decay.

TRANSURANIC ELEMENTS (6, 7)

The recycle of power reactor fuel will result in the production of transuranic elements which are only less important as hazards in long-decayed waste than are strontium 90 and cesium 137. The most important of these heavy elements are americium 241, plutonium 238, 239, and 240, and curium 242. The quantities of these elements expected to be accumulated in the waste system by 1990 are:

Americium 241	4.8 by 10^5 curies.
Plutonium 238	3.3 by 10^5 curies.
Plutonium 239 and 240	1.2 by 10^5 curies.
Curium 242	1.4 by 10^6 curies.

They become important in waste from power reactor fuel because the fissionable material is recycled, with resulting buildup of the higher isotopes. Plutonium 238 is formed by successive neutron capture by uranium 235 and 236 and neptunium 237. Plutonium 239 is used as a fuel itself, and captures a neutron to produce plutonium 240. Americium 241 results from plutonium 240 capturing a neutron and decaying to americium 241. Curium 242 arises from americium 241 capturing a neutron and decaying to curium 242.

The quantities of plutonium isotopes shown above assume a 0.1 percent plutonium loss in fuel reprocessing. The chemical behavior of americium and curium is similar to that of the rare earth fission products, and they appear in the waste stream. Any program aimed at treating wastes for hazards removal prior to discharge to the environment must take cognizance of the transuranic elements as well as the fission products. Since the process chemistry of americium and curium in waste solutions has been relatively little studied, the problem may be a formidable one.

LOW-ACTIVITY WASTES

Fuel reprocessing operations produce large quantities of low-activity wastes. These wastes are now dispersed to the environment in remote areas, but such dispersal will probably not be feasible in heavily populated sections where central fuel reprocessing plants are likely to be located. If these wastes are stored, their large volume will adversely affect the economics of nuclear power. It is hoped that it will be possible to recycle essentially all this low-activity waste within the chemical plant, thereby minimizing the volume to be disposed of.

DISTRIBUTION OF RADIOACTIVITY IN THE POWER REACTOR COMPLEX

A study has been made of the distribution of accumulated radioactivity over the power reactor complex, which consists of the reactor, decay storage prior to chemical processing, and, finally, waste storage (5, 7). The results show that the waste disposal system will contain the bulk of the long-lived, hazardous radioactivity produced by the power industry. In the case of cesium 137 and strontium 90 expected to accumulate by the year 2000, about 4 percent will be in the reactors, 7 percent in fuel decay storage, 1 percent in the chemical reprocessing plant, and 88 percent in the waste-disposal system.

ECONOMIC IMPLICATIONS OF WASTE DISPOSAL (8–10)

Tank storage: In order to assess the significance of waste disposal cost in the production of economic nuclear power, let us assume that power is to be produced for 8 mills per kilowatt-hour of electricity and that 2 percent of this cost, or 0.16 mill per kilowatt-hour may be allocated to waste disposal. Figure 4 shows the allowable waste disposal cost as a function of fuel burnup and waste volume per ton of

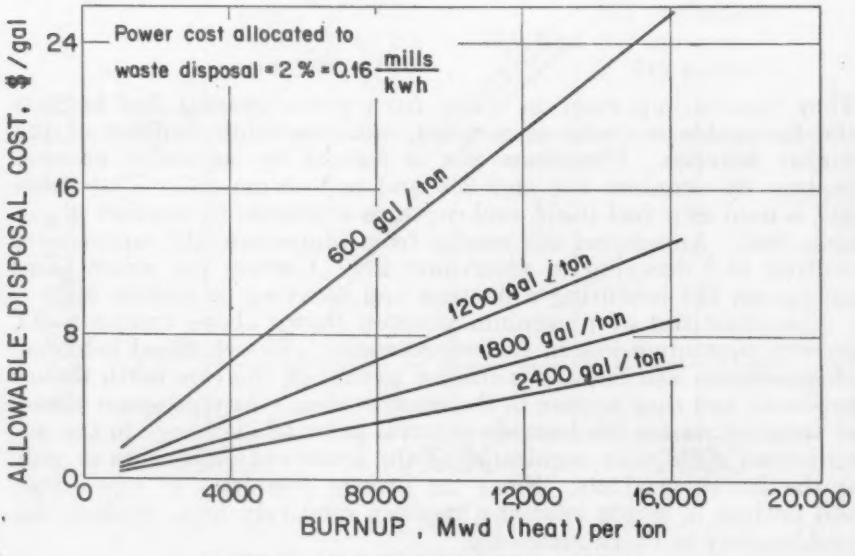

FIGURE 4.—Allowable waste disposal cost.

fuel. It now seems likely that the expected power reactor fuels will achieve burnups of about 10,000 megawatt-days per ton and that the volume of high-level waste produced will be 1,200 to 1,300 gallons per ton. Under this set of assumptions, we can afford to spend about $8 per gallon for waste disposal.

In current practice, tank storage of high-level, neutralized waste from aluminum-clad uranium fuel varies from 40 cents to $1 per gallon, depending on the tank design (3). Therefore, power cost allocated to high-level waste storage may be about 0.02 mill per kilowatt-hour. It seems apparent that the difference between the current cost of high-level waste storage and the amount which may be allocated to waste disposal, namely 0.14 mill per kilowatt-hour, is more than sufficient to treat and dispose of the low-level waste.

It is obvious that the initial cost of waste storage in tanks does not contribute greatly to the cost of nuclear power. However, since steel tanks have a finite life and will have to be replaced many times, it is not now possible to assign a realistic cost to tank storage as a method of ultimate waste disposal.

IMPACT OF RADIOACTIVE WASTES ON FUEL REPROCESSING PLANT LOCATION

The cost of shipping liquid waste from a central fuel reprocessing plant to an ultimate storage site is not prohibitive. However, this practice requires the transit of such large quantities of waste that it raises questions of safety. In order to achieve freedom in locating fuel reprocessing plants in the optimum location, attention must be given to possibilities of converting wastes to a safe form for storage and to safeguarding against atmospheric contamination.

Power reactor fuel reprocessing studies indicate that attaining reprocessing charges which are consistent with economic nuclear power requires a minimum plant capacity of 6 tons of fuel per day (11, 12). Thus, a central reprocessing plant will service many reactors. In order to minimize fuel shipping costs it is desirable to locate the plant centrally with respect to the reactors. Two sites appear attractive for early location of a fuel reprocessing plant: one on the Ohio River and the other on the Great Lakes, perhaps between Cleveland and Buffalo. Both these areas are heavily populated and the hazard associated with storage of large quantities of fission products has stimulated consideration of waste shipment to permanent disposal in salt beds. The magnitude of the waste shipment problem anticipated in the year 1980 is shown by the following values:

	1980	2000
Number of carriers in transit	505	3,270
Probable carrier weights, tons	6	6
Volume of waste in transit, gallons	227,000	1,470,000
Amount of fission products in transit, curies	1.5×10^8	9.8×10^8

In this study, (13) it was assumed that wastes were shipped a distance of 500 miles after 2,000 days' decay, the decay time which minimized the combined decay storage costs and shipping costs. Under these conditions, the total cost for decay storage and shipping 500 miles was about $1.80 per gallon, or 0.04 mill per kilowatt-hour of elec-

tricity. This liquid waste disposal requires transit of such large quantities of waste that it raises questions of safety. In order to achieve freedom in locating fuel reprocessing plants in the optimum location, attention must be given to the possibilities of converting waste to a safe form for storage or shipping.

CONCLUSIONS

The prospective nuclear power industry will produce large quantities of spent fuel, containing fission products. It is economically desirable to recover the fissionable and fertile material from this fuel in central reprocessing plants which should be located near the centers of power consumption and population. This will necessitate developing improved methods for removing iodine from off-gases, and over the long-term future, krypton 85, also.

Tank storage of power reactor fuel wastes will be complicated by their content of dissolved zirconium, stainless steel, and fluoride ion. The initial cost, 0.02 mill per kilowatt-hour, of storing power reactor wastes in tanks is not economically significant, but this storage method does not represent a permanent solution to the problem. In addition, storage of large quantities of liquid waste near centers of population must be examined from a safety standpoint. The alternative of shipping liquid wastes from the fuel reprocessing site to another location for disposal is not prohibitive costwise, but involves movement of such large quantities of radioactivity that questions of safety are raised.

Research and development on waste disposal methods for the power reactor industry should be concentrated on devising improved methods for discontaminating off-gases and on conversion of wastes to a more suitable form than the liquid for storage. The indications are that, with adequate development, these objectives can be attained at a cost commensurate with economic nuclear power. The disposal procedures resulting are expected to be sufficiently safe to make waste disposal nonrestrictive in the location of fuel reprocessing plants.

I thank you.

(The references in the foregoing statement are as follows:)

(1) J. A. Lane, "Determining Nuclear Fuel Requirements for Large-Scale Industrial Power," Nucleonics 12(10), 65 (1954).

(2) E. D. Arnold, A. T. Gresky, J. O. Blomeke, W. DeLaguna, E. D. Struxness, "Compilation and Analysis of Waste Disposal Information," ORNL–CF–57–2–20 (Feb. 11, 1957).

(3) O. H. Pilkey et al., "The Storage of High Level Radioactive Wastes: Design and Operating Experience in the U.S.," Second International Conference on Peaceful Uses of Atomic Energy, Geneva, 1954. Paper 389.

(4) J. O. Blomeke, M. F. Todd, "U–235 Fission Product Production as a Function of Thermal Neutron Flux, Irradiation Time and Decay Time," ORNL–2127 (Oct. 1, 1958).

(5) E. D. Arnold, unpublished calculations and estimates, Oak Ridge National Laboratory.

(6) J. O. Blomeke, "The Buildup of Heavy Isotopes During Thermal Neutron Irradiation of Uranium Reactor Fuels," ORNL–2126 (Dec. 20, 1956).

(7) E. D. Arnold, "Effect of Recycle of Uranium Through Reactor and Gaseous Diffusion Plant on Buildup of Important Transmutation Products in Irradiated Power Reactor Fuels," ORNL–2104 (Aug. 21, 1956).

(8) H. R. Zeitlin, J. W. Ullmann, "Economics of Waste Disposal," Nucleonics 15(1), 58–62 (1957).

(9) E. D. Arnold, "Certain Aspects of the Economics and Hazard Potential of Radioactive Waste Disposal," paper presented June 11, 1957, American Nuclear Society, Pittsburgh.
(10) H. R. Zeitlin, "Economic Requirements for Radioactive Waste Disposal in a Nuclear Power Economy," ORNL–CF–55–6–152 (June 9, 1955).
(11) F. L. Culler, "General Economics of Chemical Reprocessing for Solvent Extraction Processing," ORNL–CF–57–4–100 (May 20, 1957).
(12) C. E. Guthrie, "Radiochemical Reprocessing Costs in an Expanding Nuclear Economy," ORNL–CF–58–11–69 (Nov. 20, 1958).
(13) F. L. Culler, "Comments on the Transportation of Irradiated Fuel and Radioactive Wastes," ORNL–CF–57–5–24, pp. 4–16 (May 6, 1957).

Representative HOLIFIELD. Thank you, Mr. Bruce.

Mr. TOLL. Dr. Bruce, the other witnesses have indicated that they believe, or some of them at least have assumed, that the chemical processing plants would be located far removed from populated areas. As I understand your paper, you believe that if the gaseous discharge and waste disposal problems could be brought under control, it would be best to locate these plants near populated areas in order to cut down on transportation costs.

Dr. BRUCE. This is my feeling. However, if one does not find an adequate solution to the problem of safely storing the very large quantities of waste that will be produced, it may be necessary to locate the chemical plants in a remote area. The disadvantage of this is that it involves the shipping of the fuel elements for rather extensive distances, and this, of course, contributes to the cost of nuclear power. On the other hand, if one examines the cost of shipping fuel elements, just as we have here examined the cost of shipping waste, it can be shown that the cost of shipping fuel elements is not a really large item in the cost of nuclear power.

So I think that one would not make a very, very strong argument for locating the chemical plants in centers of high population density. However, there is incentive to do so if all the problems attendant to this are solved.

Dr. RODGER. Dr. Bruce, you said this in your paper. I think it would be well to emphasize that in obtaining the value of 0.02 mill per kilowatt-hour, you were assuming a burnup of 10,000 megawatt-days per ton. On the basis that we are operating today, these numbers would be at least a factor of 10 higher. In some respects we are solving the waste disposal problem by solving the fuel element problem.

Dr. BRUCE. Yes.

Dr. RODGER. In line with your comment concerning the cost of the transportation, when you come out with the number of 0.04 mill per kilowatt-hour for shipping and 0.02 mill per kilowatt-hour for storage, one may probably conclude that, even shipping as short a distance as 500 miles, the cost of shipping might be twice the cost of waste disposal.

Dr. BRUCE. This is true if you are willing to consider tank storage as waste disposal. If, on the other hand, one assumes that the cost of tank storage should be regarded as a dollar per gallon per year figure, which goes on and on and on for many hundreds of years, then the cost of tank storage may be a very large item.

Dr. RODGER. Yes, the allowable figure you mention of $8 per gallon for the total cost of the entire operation would roughly be eight times this amount.

Dr. BRUCE. Yes.

Dr. RODGER. Thank you.

Mr. TOLL. If one puts primary emphasis on safety, as I suppose all of us would, which would you say was the greatest safety hazard, the shipping of solid fuel elements, or the location of chemical processing plants involving high-level liquid wastes in highly populated areas?

Dr. BRUCE. I would say that the greater hazard is associated with the fuel reprocessing plant located in a highly populated area and storing liquid wastes. I think for this reason that there is great incentive to develop safer methods for storing these wastes than in the form of liquids. It is possible to visualize many schemes which appear to be extremely safe for handling these materials even in highly populated areas.

Representative HOLIFIELD. Thank you, Mr. Bruce.

The next witness is Mr. H. M. Parker of Hanford.

STATEMENT OF H. M. PARKER,[2] MANAGER, HANFORD LABORATORIES, RICHLAND, WASH.

Mr. PARKER. Mr. Chairman, my name is H. M. Parker. I am employed by the General Electric Co. at the Hanford atomic products operation in Richland, Wash., as manager of the Hanford Laboratories.

In an earlier part of these hearings I was privileged to present to you an account of the waste management operations of the General Electric Co. at the Hanford plant.

In the present case it is intended that these speculations on the future problems in waste disposal be primarily personal views, not necessarily reflecting the best balanced judgment of any organization.

The first requirement is to categorize the future magnitude of the wastes. Various projections of the growth of a nuclear power industry have been made. I shall use that of J.A. Lane ("Determining Nuclear Fuel Requirements for Large-Scale Industrial Power," Nucleonics (12(10), 65, 1954) and the additional calculations made by F. L. Culler, Jr., and S. W. McLain for the Committee on Disposal and Dispersal of Radioactive Wastes of the National Academy of Sciences—National Research Council ("Status Report on the Disposal of Radioactive Wastes," ORNL–CF57–3–114 rev., June 25, 1957). All these numbers will be in agreement with those just presented by Mr. Bruce.

The projection gives—

For 1970, nuclear power (heat) : 33,000 MW.
For 2000, nuclear power (heat) : 700,000 MW.
For 1970, high activity waste: 5 to 20 million gallons.*
For 2000, high activity waste: 200 to 1,000 million gallons.*

To the present time, the Hanford plant has stored about 50 million gallons of high-level waste. We can assume, for the present purposes, that high-activity wastes and high-level wastes in these two contexts, while far from identical, are of the same general nature.

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[2] See p. 159.

* Based on 820 gallons per ton of spent fuel at exposures of 2,000 to 10,000 megawatt-days per ton.

Assuming the accuracy of these predictions, the total national storage requirement originating from nuclear power will be only 10 percent to 40 percent of Hanford's demonstrated storage by 1970.

Probably it will be between 20 and 25 years before the national storage demand from nuclear power equals the present demand at Hanford. This is greater than the length of time during which a significant radioactive waste disposal problem has existed.

Knowledge of the factors involved in waste disposal has grown significantly in the last 10 years. I am confident that American science and technology can easily keep pace with the need during the next quarter century.

By the year A.D. 2000 the national storage requirement from nuclear power is projected to be 4 to 20 times that of Hanford today. The storage and manipulation of such a quantity in tanks at one location or at a number of carefully selected locations would be quite feasible.

It is usually assumed that control of the main mass of fission-product wastes is the determining feature in waste disposal. If this is correct, it follows that there is no insuperable problem in control by storage alone for the next 40 years. That superior methods, such as binding of wastes in ceramics, or other more permanent forms of retention, will not be in place, and in place economically, in this period, is inconceivable.

In this broad look at the storage of the main radioactive wastes, details of changes in those wastes brought about by changes in reactor technology are relatively insignificant. Some of the likely changes may help and others hinder disposal. Foreseeable changes in chemical reprocessing are generally in the direction of smaller volumes of waste, reduction to dry, nonleaching, nonpowdering forms, and eventually to elimination of aqueous intermediate forms. All these should tend to alleviate the waste management problems.

The potentially significant waste disposal problems up to A.D. 2000 become—

1. Incidental, more or less regular, releases of radio-active material to the environs during fuel processing, reactor operation, or chemical reprocessing.
2. Incidental leaching or relocation of small fractions of major wastes nominally immobilized.
3. Substantial accidental, sporadic, or irregular releases of various kinds.

We shall consider only the first category. The estimated increase in storage demand is in the range of 4 to 20. Consider the effects of multiplication factors of 5, 20, and 50 applied to the present Hanford system.

For the release of I^{131}, the emitted quantity would have to be maintained at about the present level either by improving the removal equipment or by increasing fuel cooling time—45 days takes care of the full factor of 50.

For the radioactive noble gases, as mentioned earlier by Mr. Bruce, trapping and storing may be necessary to avoid undue increase in the worldwide background. These technologies are straightforward. We shall use the language "straightforward technology" when the

technological success is assured and the cost is less than 5 percent of the total cost of the installation. This avoids detailed calculation of engineering costs that will probably never be used.

The release of discrete radioactive particles to the atmosphere has not yet been eliminated entirely. Present filtration allows about one particle out of a thousand to escape. To increase the efficiency of removal tenfold would be technologically straightforward in the above special sense. Beyond that point increased releases might be needed. These would still be small compared with currently accepted quantities arising from fallout.

Substantial quantities of liquid radioactive wastes in the classes known as intermediate and low-level, have been released to the ground at Hanford. Although the assurance with which such material is kept out of the effective environment is less than if it were tanked, there is no evidence that such disposal under Hanford conditions is unsuitable.

Those connected with this form of disposal judge that a fivefold, twentyfold or even fiftyfold increase in releases of this character would present no major problem. Nevertheless, increases at the upper end of this range would not be generally acceptable without substantial demonstration of the maintained effectiveness of the system.

In summary, the adequate management of wastes in A.D. 2000 by simple extension of present methods is not out of the question. The incidental releases during various processing stages will, however, require careful and continued study to assure that the combined releases are not excessive.

Here I should like to refer back to the section entitled "Transfer Mechanisms of Released Wastes" in part I of the material filed for the record by the staff of the Hanford atomic products operation under Topic III—Waste Management Operations. This is repeated for convenience in slightly modified form as an appendix to this paper.

(The appendix referred to is as follows:)

APPENDIX

EXPOSURE PATHWAYS OF RELEASED RADIOACTIVE WASTES

When radioactive material is released to the environment there is a complex web by which the material is transmitted from one component, animate or inanimate, to another. At each point in the web or chain, man or other life forms may suffer some degree of radiation exposure. The combined effect of all relevant pathways has to be determined.

The following schematic presentation of the principal pathways follows that given in Part I of the material filed for the record under Topic III by the staff of the Hanford Atomic Products Operation. Some notes are added on pathways involving the ocean and marine organisms; these were irrelevant in the context of our Topic III presentation.

Those chains likely to make the major contributions to total exposure are identified by double asterisks. Single asterisks identify the next group of contributing chains. This identification is intended to be approximate only, as an average across a postulated nuclear power industry.

The situation around any particular site must be determined experimentally for that site.

Case 1. -- Radioactive Wastes in Rivers, Streams, Lakes, or Oceans

1.1. - Hazards related to use as drinking water

** 1.1.1 Irradiation of gastrointestinal tract of man or animals.

* --.2 Transfer of radioactive materials to blood stream and irradiation of blood.

** --.3 Transfer to critical organ of deposition (e.g. liver, kidney, bone, or thyroid) as determined by the specific radioelement and its chemical and physical form.

** 1.1.4 Irradiation of reproductive organs from materials passing through the body, deposited in specific organs, or in rare cases deposited in the reproductive organs.

--.5 Retention of filtrable radioactive materials in water purification systems, leading to external irradiation.

* --.6 Retention of non-filtrable materials in ion-exchange systems (e.g., water softeners) or in flocculent deposits in municipal water systems.

--.7 Scaly deposits of radioactive materials from boiling hard water.

--.8 Uptake by foods washed or cooked in domestic water.

1.2.- Hazards arising from immersion in the water

1.2.1 External irradiation from the ambient water while swimming.

--.2 Additional irradiation from materials attached to mud, rocks, piling, aquatic plants; or, in fact, any surface.

--.3 Irradiation from materials deposited on the skin or clothing.

* --.4 Swallowing of water, equivalent to Case 1.1.-

--.5 Irradiation similar to Cases 1.2.1 and 1.2.2 from use of sanitary water.

1.3.- Hazards arising from close approach to the water

1.3.1 External irradiation from river, lake, reservoir, or ocean.

1.3.2 External irradiation from mud, sand, vegetation, or salt deposits near the shore, on beaches, or tide flats.

--.3 External irradiation from water while boating, water skiing, fishermen wading, etc.

--.4 Augmentation of 1.3.3 by radioactive deposits on the boat structure, fishing gear, navigation locks, etc.

--.5 Inhalation of contaminated spray. (→ Case 2.--)‡

--.6 Deposition of spray on skin or clothing. (→ Case 2.--)

--.7 Similar situations from domestic use of sanitary water.

1.4.- Hazards arising from irrigation

1.4.1 Irradiation from water in canals or ditches

--.2 Retention of radioactive material in the surfaces of canals or ditches, persisting in the absence of irrigation water.

* --.3 Atmospheric pickup of radioactive materials from these cases or from irrigated soils. (→ Case 2.--)

--.4 Direct spraying of plant surfaces with contaminated water by overhead irrigation.

** --.5 Immediate uptake of radioactive materials by plant roots from the water or delayed uptake following retention in the soil, in either case leading to incorporation in the plant structure; this can be followed by direct irradiation from the crops, or ingestion of the radioactive material by man or range animals.

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‡ Symbols such as (→ Case 2.--) mean that this case leads also to some of the consequences of another case, such as Case 2.-- in this example.

1.4.6 Further transfer and dissemination of radioactive materials by birds or insects.

--.7 Transfer to the atmosphere by burning of crops.

* --.8 Radioactive milk, eggs, or meat from range animals or poultry fed as in 1.4.5.

1.5.- Hazards arising from biological chains in fresh water

*1.5.1 Pronounced uptake of radioactive materials by attached algae or by plankton (free floating organisms).

* --.2 Transfer to snails, insect larvae, etc. directly or from 1.5.1.

* --.3 Transfer to higher forms including edible fish.

** --.4 Ingestion of fish by man.

* --.5 Transfer to aquatic plants

* --.6 Transfer to waterfowl from aquatic plants, insects, etc.

* --.7 Ingestion of contaminated fowl by man.

--.8 More exotic food chains such as ingestion of contaminated insects by frogs and the frog's legs by man.

--.9 Mass emergence of contaminated ephemeral insects, their infestation of dwellings, and consumption by birds.

--.10 Secondary contamination of bird nests and the like from excreta.

--.11 Absorption of radioisotopes into fish eggs.

1.5.12 Perpetual recycling of isotopes through biological chains through death and decomposition of organisms.

In any of these transfers, the transmitter as well as the receptor will receive radiation exposure.

1.6.- Hazards arising from biological chains in the ocean

*1.6.1 Uptake similar to 1.5.1 but including seaweeds.

--.2 Transfer to invertebrates as in 1.5.2 but shellfish are of greater significance.

Note: Although marine organisms pick up radioactive materials by processes which are similar to those for fresh water forms, the degree of biological concentration for any particular radioelement may be considerably different. For example, radio-cesium is highly concentrated in fresh water organism but apparently is not concentrated in substantial amounts from sea water.

* --.3 Transfers to fish, birds, and other animals as indicated in 1.5.3, 1.5.5, 1.5.6, 1.5.10, 1.5.11, and 1.5.12.

* --.4 Ingestion of radioactive materials by man from eating seaweed, shellfish, fish, or fowl.

1.7.- Hazards related to industrial processes

1.7.1 Direct irradiation equivalent to 1.3.-

--.2 Preferential adsorption of radioactive materials on food products during washing.

1.8.- Hazards related to sewage disposal

1.8.1 Accumulation of radioactive materials in drains later requiring service.

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1.8.2 Accumulation in sludge and filter beds.

--.3 Accumulation in bacteria, affecting their function.

* --.4 Concentration and return to agriculture via fertilizers.

Case 2. -- Radioactive Wastes Released to the Atmosphere

2.1.- Hazards related to inhalation

2.1.1 Direct irradiation of the lung due to radioactivity of the contained air.

--.2 Solubility of radioactive gases or vapors and transfer to the blood and the specific organs of deposition. (→ 1.1.2 and 1.1.3)

--.3 Deposition of radioactive particles on the surfaces of lung, bronchi, and upper respiratory passages, giving intense local irradiation.

--.4 Solubility of some of these particles. (→ 2.1.2)

** --.5 Transfer of particles to lymph nodes of the lung.

--.6 Removal of particles by ciliary action, leading to ingestion. (→ 1.1.-)

2.2.- Hazards from radioactive material in ambient air

* 2.2.1 External irradiation of the human or animal body, or plant tissue.

2.3.- Hazards related to deposition of materials

2.3.1 Irradiation from deposition on skin of man or animals, or on clothing.

--.2 Transfer of radioactive materials from other surfaces to skin.

--.3 Secondary transfers to food, cigarettes, etc., leading to ingestion.

** 2.3.4 Deposition on crops, followed by transfer to man or animals.

** --.5 Secondary transfer of the radioactive materials through use of products such as milk, eggs, and meat.

* --.6 Deposition on soil. (→ 1.4.5)

--.7 Deposition on bodies of water. (→ Case 1.--)

--.8 External irradiation from deposited materials.

--.9 Transfer from one animal to a predator animal.

--.10 Washing by rain or floods into water systems. (→ Case 1.--)

* --.11 Real fallout of individual highly active particles or flakes onto hair or skin.

Case 3. -- Ground Disposal of Liquid Wastes

3.1.-Hazards at the disposal site

* 3.1.1 If to surface ponds, the hazards are repetitious of many parts of Case 1.--, particularly 1.3.- (The ponds may be particularly attractive to wildfowl.)

* --.2 If contaminated surface is dried, transfer of radioactive materials to atmosphere by wind pickup of soil. (→ Case 2.--)

--.3 Direct irradiation from a source just below ground.

--.4 Seepage of radioactive gases or vapors from below ground.

* --.5 Flash floods removing superficial active materials. (→ Case 1.--)

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3.2.- Hazards resulting from underground travel

3.2.1 Return of radioactive material to plant life from deep-rooted plants.

--.2 Subsequent leaching out of radioactive material apparently fixed in soil.

--.3 Chemical reactions releasing materials as incompatible wastes are later added.

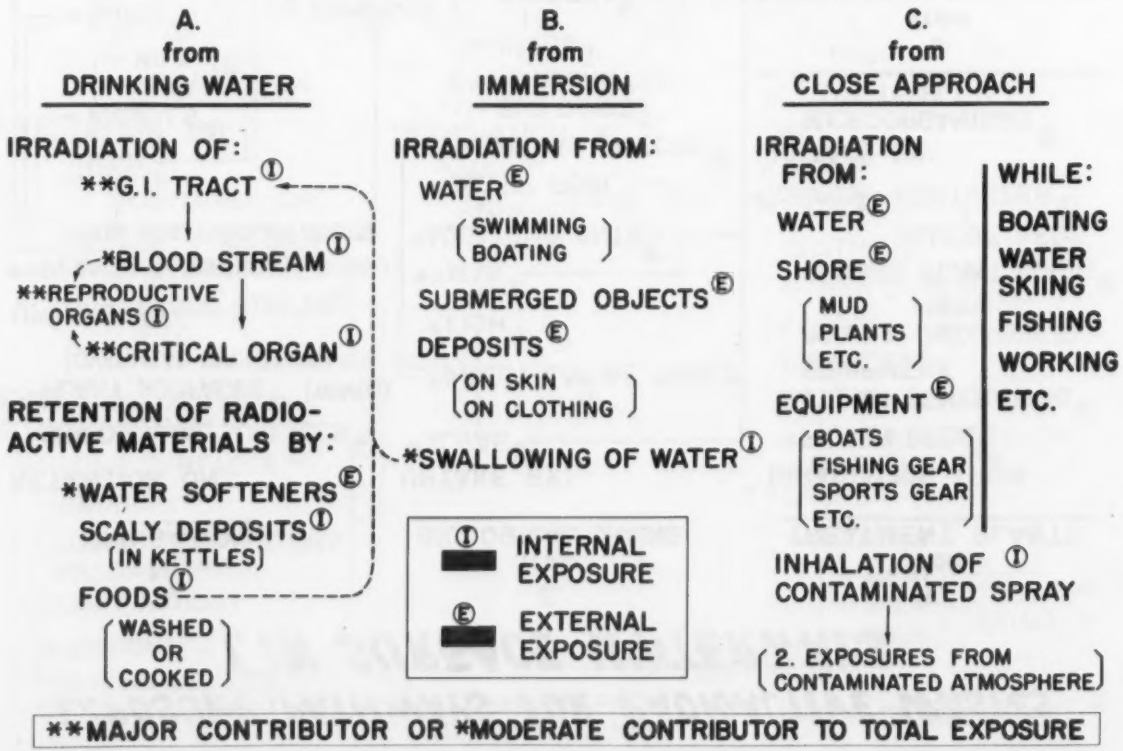
--.4 Slow fall of wastes under gravity when apparently safely separated from the ground water.

* --.5 Entry into ground water and subsequent recovery from wells (→ Case 1.--)

* --.6 Underground travel to springs or rivers. (→ Case 1.--)

EXPOSURE PATHWAYS FOR RADIOACTIVE WASTES

1. IN SURFACE WATERWAYS



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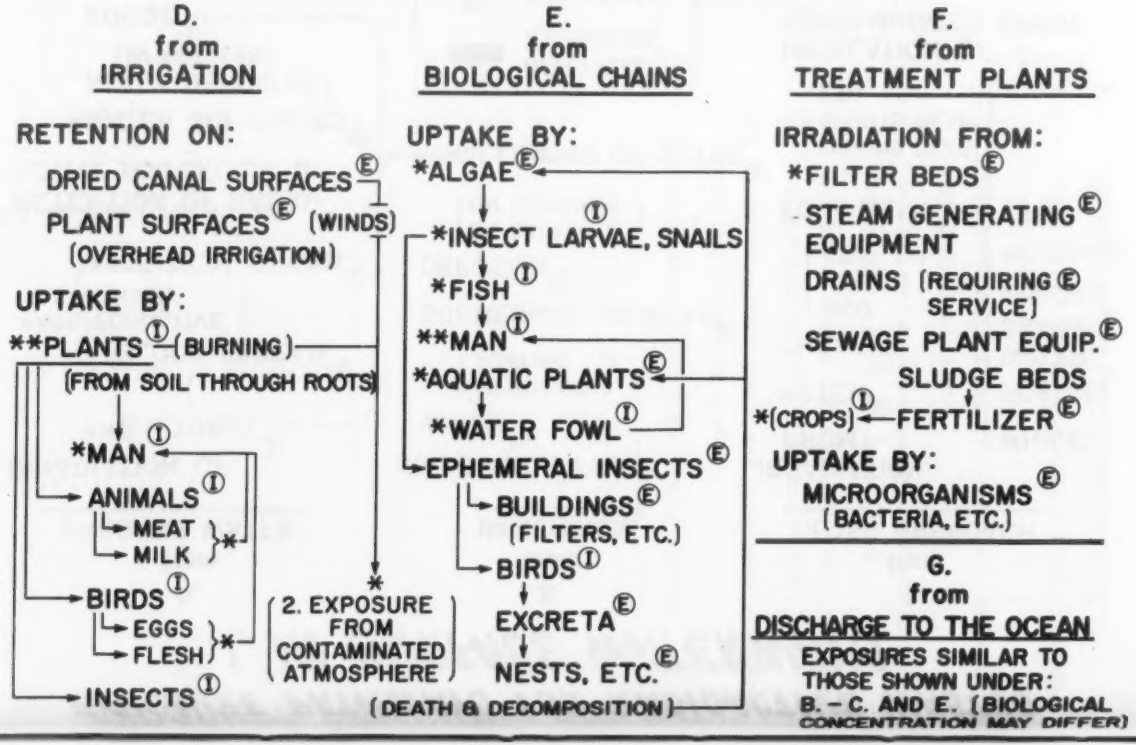
EXPOSURE PATHWAYS FOR RADIOACTIVE WASTES
I. IN SURFACE WATERWAYS



EXPOSURE PATHWAYS FOR RADIOACTIVE WASTES

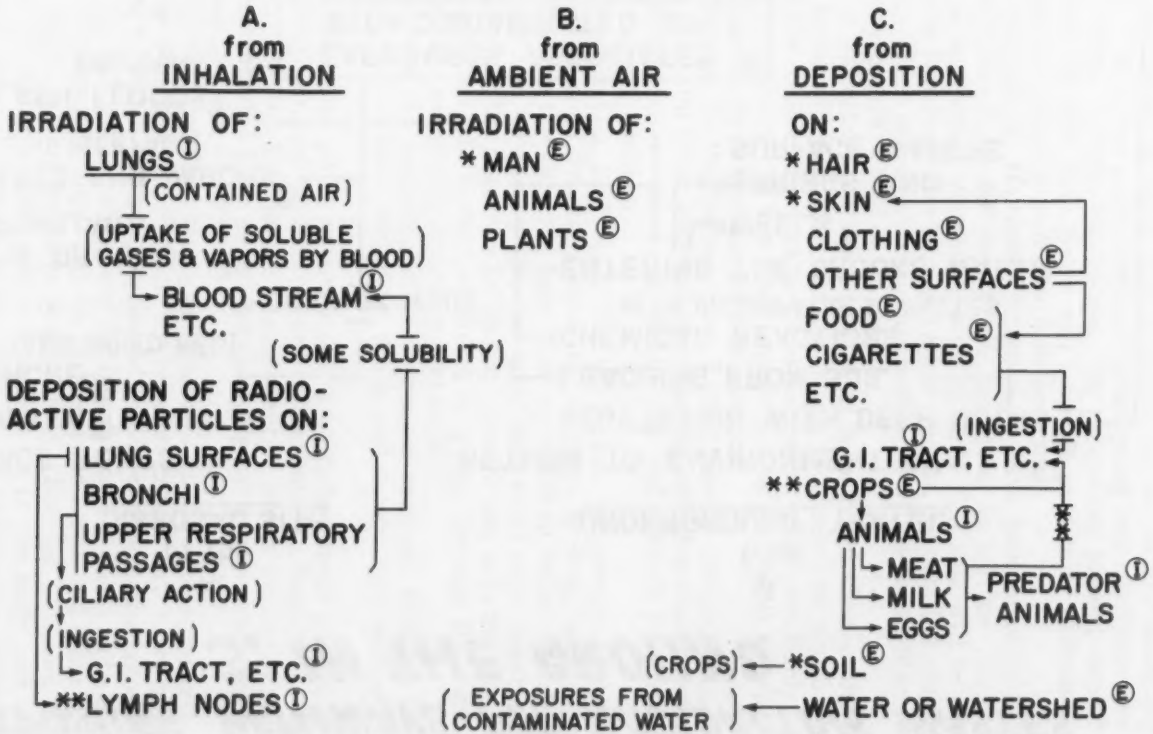
→INSECTS Ⓔ

(DEATH & DECOMPOSITION)

B. – C. AND E. (BIOLOGICAL CONCENTRATION MAY DIFFER)

EXPOSURE PATHWAYS FOR RADIOACTIVE WASTES

2. IN THE ATMOSPHERE



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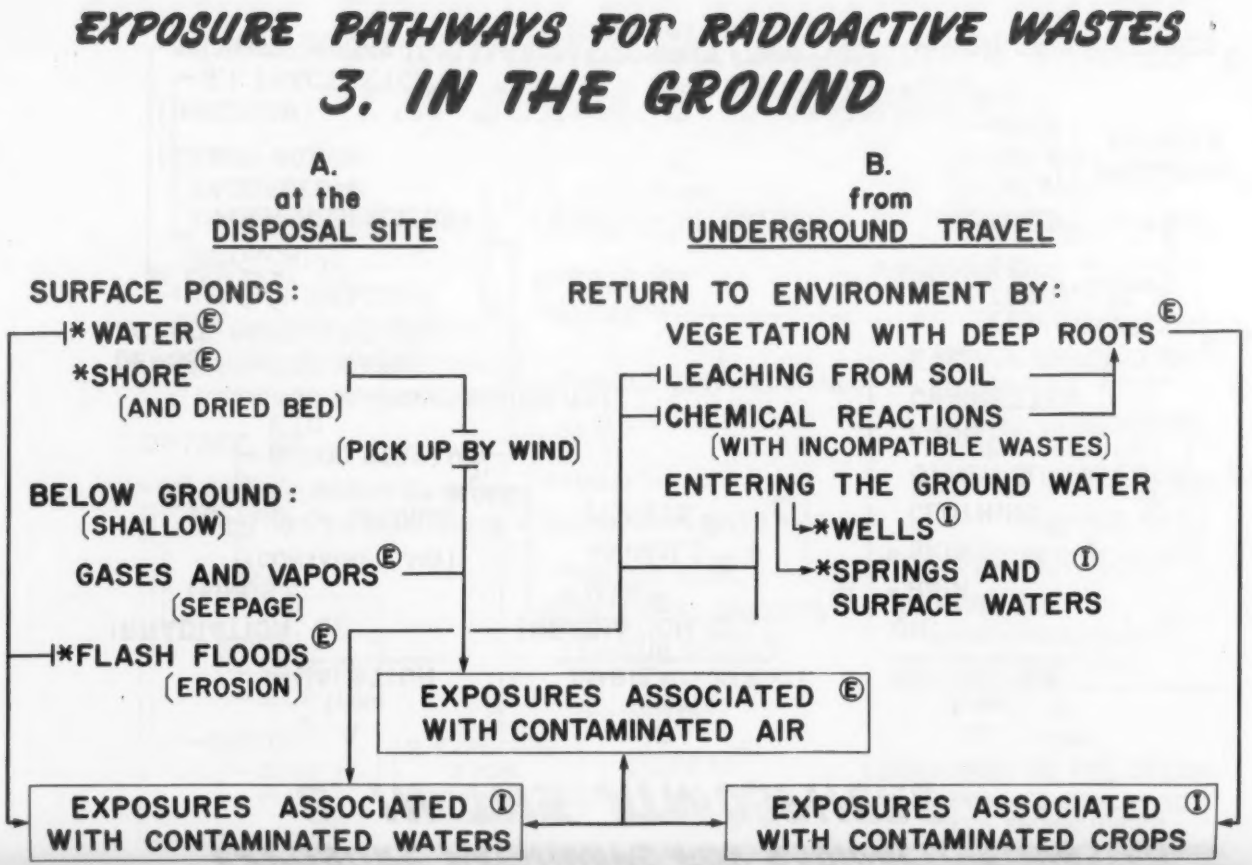
EXPOSURE PATHWAYS FOR RADIOACTIVE WASTES
3. IN THE GROUND
A.
at the
DISPOSAL SITE
B.
from
UNDERGROUND TRAVEL
SURFACE PONDS:
*WATER (E)
*SHORE (E)
(AND DRIED BED)
(PICK UP BY WIND)
BELOW GROUND:
(SHALLOW)
GASES AND VAPORS (E)
(SEEPAGE)
*FLASH FLOODS (E)
(EROSION)
RETURN TO ENVIRONMENT BY:
VEGETATION WITH DEEP ROOTS (E)
LEACHING FROM SOIL
CHEMICAL REACTIONS
(WITH INCOMPATIBLE WASTES)
ENTERING THE GROUND WATER
*WELLS (I)
*SPRINGS AND SURFACE WATERS (I)
EXPOSURES ASSOCIATED WITH CONTAMINATED AIR (E)
EXPOSURES ASSOCIATED WITH CONTAMINATED WATERS (I)
EXPOSURES ASSOCIATED WITH CONTAMINATED CROPS (I)

Mr. PARKER. These charts (as given in the preceding appendix) are taken in the order in which they appeared in our earlier testimony and so the first refers to radioactive wastes going into environmental pathways from surface waterways. In projecting present experience to that of 2000 A.D. there will be some particular differences which cannot all be condensed into these diagrams.

As an example, at Hanford currently we have been substantially concerned with releases of reactor cooling water to a river. In the overall nuclear power technology it is unlikely that this will be one of the major features. For this and other reasons, the detailed projection of just which individual exposure pathways will be the significant ones by the year 2000 would be too bold.

We have indicated on these charts what is no better than a rough guess in terms of these asterisks (pointing to charts) in which we represent the double asterisks as those pathways which are likely to be major contributors to the total exposure, and the single asterisks those that are likely to be moderate contributors.

This morning, in the limited time, I shall merely categorize these charts by title and draw your attention to the complex chains in the environment which we see as the really key and difficult portion of the waste disposal problem.

In the first one (pp. 2367 and 2368), the surface waterways, we have the complex A from drinking water, the complex B from immersion of individuals in the water, C from close approach to bodies of water, and continued on the next chart, still in surface waterways, the transfer to plants and the like from irrigation use of water as in Section D, and transfer from the biological chains in rivers, through the algae, insects, fish, and eventually to man in Section E.

In fact, here the recycling of this released material through the whole ecological system, including death and decay, may be highly significant.

Introduction into man's environment through treatment plants, filter beds, fertilizer, and back into crops is illustrated in Section F. In this corner (Section G) there is a brief mention of the similar system in the ocean which I have added here although it was not relevant in the original testimony of Part III.

The next chart (p. 2369) shows a similar situation for exposure pathways in the atmosphere. First, in the inhalation pathway A, we have the system in which the introduction of radioactive particles into the lung and probably ultimately into the lymph nodes is likely to be one of the determining factors. The air that is around man and other life forms is shown as an exposure source in B. Section C shows the deposition of material from the atmosphere on to the body and the ground, and especially on to crops. Cycling of radioactive material through crops, animals, animal products and around to man is shown. Other chains lead from soil into crops. From deposition into water and back to the previous channels (p. 2370) for surface waterways is just one way in which *all* the pathways on *all* the charts tend to interlock.

Finally, chart 3 shows the pathways of wastes disposed into the ground. The local things at the disposal site (Section A) we believe are quite controllable. The possibilities of unpredicted or unwarranted underground travel and return of the material to man's environment, such as by vegetation with deep roots, leaching from soil,

eventual access to water through wells and the like, are sketched in section B.

In specific cases, quite devious exposure pathways, ones not currently tabulated, or the use of special foods foreign to our diets (seaweed, insects, as examples) may be the controlling factors.

If Hanford experience is a guide, the total radiation exposure will be composed of many different contributions. This greatly complicates dose measurement or calculation, interpretation of the radiological significance, and prediction of the future status at higher outputs.

In addition, if there are multiple release sites, the exposure patterns around them will eventually overlap. Worldwide fallout also adds to the local patterns.

Not the least significant of the uncertainties is that in the acceptable permissible limits. Reduction of such limits by a factor of 10 by administrative fiat can be a prompt process; growth of processing capability by a factor of 10 is comparatively slow and predictable. Yet the net result on the waste management is much the same.

As a condensed forecast of the situation to 2000 A.D.:

1. The expanded use of atomic energy need not be inhibited at all by the radioactive waste disposal problem, unless there should be concurrently a drastic reduction in permissible radiation exposures.

2. Appropriate improvements in control of wastes appear to be technologically straightforward, in the sense that feasible methods at reasonable cost are known.

3. Continuous examination of the exposure pathways around each principal release site is needed.

4. Extensive research into three areas of the radiological sciences will be needed. These are:

(*a*) The mechanisms of transmittal of radioactive materials through numerous environmental pathways.

(*b*) The significant radiation exposure to man and other life forms as receptors from these channels.

(*c*) Acceptable limits of exposure of man and other life forms as individual receptors and as components of a genetic pool.

Mr. Chairman, that concludes the matters which I wish to present.

Representative HOLIFIELD. Thank you, Mr. Parker, for your statement. Are there any questions?

Thank you, sir. Dr. Morgan, of the ORNL, will be the next witness.

STATEMENT OF K. Z. MORGAN,[3] DIRECTOR, HEALTH PHYSICS DIVISION, OAK RIDGE NATIONAL LABORATORY, OAK RIDGE, TENN.

Dr. MORGAN. Mr. Chairman, members of the committee, I will summarize and evaluate some of the environmental factors that must be considered in the disposal of radioactive wastes.

We have seen from the foregoing discussions that there has been a rapid increase in the amount of radioactive waste, an increase that parallels the growth and expansion of the nuclear energy industry and its associated operations. It has been indicated that the high

[3] See p. 428.

level liquid waste may be stored in underground tanks, but this must be considered as a temporary or interim expediency, for the integrity of tanks can be relied upon for only a few score years. It has been shown that the high level waste may be concentrated so that it may be stored permanently in isolated areas or perhaps much of it can be fabricated into large industrial sources of ionizing radiation.

Regardless of the many useful applications of these radiation sources, their ultimate disposal presents a problem and the more widespread these useful applications the more universal and ever present the associated radiation accidents and hazards. We have seen that research is underway in the AEC-supported laboratories to explore the feasibility of various methods of permanent and semipermanent disposal of high level radioactive waste. Some of these methods consist of fixation in sintered clays and sands, disposal in salt formation, and injection into deep wells, deep sea disposal, and so forth. These research studies should be continued and the results evaluated from time to time in terms of economics and the long-range radiation protection they can provide.

Radioactive waste is produced in the liquid, gaseous, and solid forms; of these, the liquid waste constitutes the principal long-range, environmental problem, for regardless of the initial form, that waste which constitutes a hazard, for the most part, has found its way by dilution or suspension into surfarce and underground waters.

Elaborate precautions are taken to minimize the air contamination problems associated with the nuclear energy industry. Most of the particulate radioactive waste is retained and barring accidents only the relatively harmless radioactive Noble gases are discharged in large quantity from reactors to the atmosphere. These gases do not present any serious problem, since the maximum permissible concentration (MPC) of Noble gases is very large and the discharge of the gases at high velocity through tall stacks provides dilution which reduces the concentration at a short distance from the point of discharge far below the maximum permissible value.

Unless there are major nuclear accidents, it is not the high level waste that introduces most of the environmental radiation problems, but rather the immense volumes of intermediate and low level radioactive waste having a concentration of a few micro-microcuries per cubic centimeter.

In the United States the values of maximum permissible exposure to ionizing radiation are established by the recommendations of the National Committee on Radiation Protection (NCRP). A similar organization, the International Commission on Radiological Protection (ICRP) sets these values for international use. These two organizations work closely together, for many of their problems, such as the disposal of radioactive waste, may be of concern to people in many countries of the world.

There are two types of exposure to ionizing radiation: (1) external, e.g., exposure from medical equipment or nuclear reactors, and (2) internal, or exposure from radioisotopes taken into the body.

In considering the environmental factors associated with the disposal of radioactive waste we are concerned primarily with internal

exposure rather than with external exposure. If the radioactive material taken into the body is relatively insoluble it is likely that most of the dose will be delivered to the lungs or to the gastrointestinal tract. If it is very soluble, a large fraction will enter the blood stream where it is deposited in the various organs of the body. Two types of elimination take place simultaneously—biological removal and radioactive decay.

There is a wide variation in the value of MPC. For example, the MPC for occupational exposure to tritium is 10 grams of tritium per million tons of water whereas the MPC for occupational exposure to strontium 90 is 0.03 gram of strontium 90 per million tons of water. This is 10 grams of wood I hold up here [holding up a small block of wood], and if it were tritium uniformly distributed in water, it would contaminate a million tons of water at the maximum permissible concentration. Strontium 90 is much more toxic. I hold up here [now holding up a small chip of wood] three one-hundredths grams of wood. If this were strontium 90 uniformly distributed in water, it likewise would contaminate a million tons of water at the maximum permissible concentration. This three one-hundredths gram of strontium 90 does not appear to be very much. Yet if deposited in our bodies it would constitute a maximum permissible body burden for 2 million persons.

For comparison, if this million tons of water, containing three one-hundredths grams of strontium 90 were normal sea water, it would contain about 6 tons of gold.

There are many uncertainties in the values of MPC because in only a few cases are human data available, and in some cases even animal data are lacking. However, the latest editions of these handbooks on internal dose contain values of MPC for about 240 radioisotopes and these data, although limited in accuracy, represent the best available at the present time. In these handbooks values are given only for occupational exposure; it is expected that in the near future similar values of MPC can be recommended for nonoccupational exposure. For the present, the interim MPC for persons living in the neighborhood of a nuclear energy operation are to be taken as 10 percent of the values for continuous occupational exposure and the values of MPC for the population at large are to be of the order of natural background. Because the average values of MPC permitted to the public are so much lower than those permitted to occupational workers, the radiation hazards of the nuclear energy industry are confined primarily to occupational workers. This is in conformance with the usual practice of restricting unique hazards of each occupation to the workers in that occupational group. Thanks to the fine job done by the health physicists, the average exposure of the occupational worker in the nuclear energy industry is only a few percent of the maximum permissible value.

At present, medical exposure constitutes essentially all the radiation exposure of the average man to man-made sources. It is not expected that the present interim radiation exposure levels for the population at large will be exceeded in the foreseeable future.

The ICRP is allowing 5 rems for planning purposes (see table 1, p. 2377) and of this 5 rems of exposure, 1½ rems is assigned to radioactive waste disposal in the environment.

If and when these levels are reached, waste disposal will contribute no more than one-third of the radiation damage received at present by the average medical sources or natural background.

Three types of damage must be considered: (1) somatic damage or direct injury to the individual, e. g., the induction of leukemia and shortening of lifespan; (2) genetic damage; and (3) indirect damage to man's environment, e.g., the effect on his food supply and the natural resources. In what follows we will limit our discussion to the ecological effects of radiation on the environment. There seems to be rather widespread feeling, especially among laboratory biologists, that since man appears to be as radiosensitive as any organism all we need to do is monitor radiation in the environment where man actually lives. Perhaps this is a dangerous oversimplification for there are many important environments such as lakes, streams, and soil where man himself is not a direct resident but upon which he depends indirectly for his food, pleasure, etc.

The possible consequences to man from contamination of these environments by radioactive materials cannot be evaluated without a study of these environments which contain populations of plants and animals that are organized into biological systems or communities, and man cannot survive independently. Communities and members of communities are dependent upon each other for their continued existence, and this type of biological organization with its complex cycles of elements, compounds, and minerals is characteristic of our entire world. Therefore, in any waste disposal program we must take into consideration not only the problem of maintaining concentrations of radioactive material in soil and water at levels considered to be safe for direct exposure to man and his children, but also we must consider the possible consequences of the escape and accumulation of small quantities of radioactive materials into the environment where long-range modifications may result in a less favorable place in which to live. In a large scale nuclear power economy where there is competition for cheaper sources of energy there will be an increasing need and demand to make use of the dilutent capacity of the environment for the economic disposal of large volumes of low level radioactive wastes. Thus, long range continuing studies must be made to assure that the levels of radioactive contamination in the environment are not kept so low as to unnecessarily impede this new industry or on the other hand permitted to become so large that the damage to the environment exceeds the overall benefits from nuclear power and its useful products.

What concerns us here is not the massive effects that might result from the accidental release of large quantities of fission products, but rather the subtle, cumulative effects that might be building up due to the slow but continuous increase in radioactive contamination of our environment. Such effects may not become manifest or may remain concealed due to other more pronounced phenomena, and may only become evident hundreds of years from now. Because of this uncertainty in our knowledge we believe that environmental research programs should be undertaken and should be specifically oriented toward the contamination problem.

There are many examples of instances in which man has accidentally or intentionally tampered with his environments and produced unde-

sirable results. When radioisotopes are released into the environment they usually enter into complex physical and biological cycles and into food chains and may exhibit unexpected movements or concentrations. It is through such mechanisms that dangerous long-lived radionuclides such as strontium 90 and cesium 137 are able to enter our food sources. These ecological food chains (as shown in figure 1) are actually circular in nature or weblike and have a limited number of links. Serious consequences could result if too many of these food links were broken. Radionuclides with long half-lives may be expected to be taken up and dispersed, and reconcentrated by ecological factors, although no one has yet followed materials through more than one cycle.

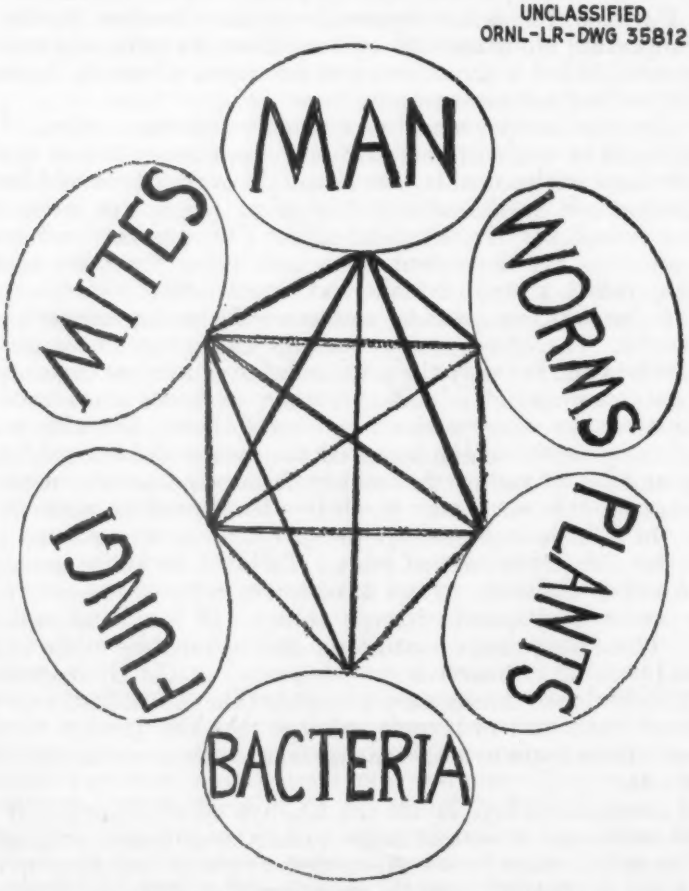

FIGURE 1.—Typical food web involving man.

Evidence of selective absorption factors for some radioisotopes as high as 100,000 has been obtained for certain organisms.

We should note that most of the studies have been made with respect to killing effect. However, there may be many other types of damage more subtle but just as damaging in their consequence to the human environment. For example, radiation may affect the germ cells such that there is a reduction in fertility.

The average dose delivered to the environment at the present time by manmade radioactive waste is very low and even in the future,

if the levels presently proposed for planning purposes (as indicated in table 1) are reached, the average dose will be no more than about 50 percent of that from natural background. However, because of selective deposition, certain elements in the food web may concentrate some of the long-lived radionuclides resulting in very high doses to some of the critical links. The end result is unknown but could be a reduction in man's food supply or a destruction of some of the natural resources.

In conclusion, I would like to point out that, although radiation hazards of new and heretofore undreamed of proportions have arisen, the nuclear energy industry through the untiring efforts and constant vigilance of the health physicists is one of the safest American industries. With continued alertness and with an increased number of long-ranch research programs, this record can be maintained and the benefits of nuclear energy can be made to dwarf by comparison any harmful effects of the radiation on man and his environment.

Representative HOLIFIELD. Thank you, Dr. Morgan.

Are there any questions of Dr. Morgan by any member of the committee or of the staff? If not, we thank you very much, sir.

Dr. MORGAN. Thank you, sir.

TABLE I

Maximum Permissible Genetic Exposure to the Population at Large*

(RBE dose in rem to the gonads to age 30)

4.5	Medical			
5.0	Other	2.0	1.5	Internal (waste disposal)
			0.5	External
			2.0	General to population at large
4.5	Background	3.0	1.0	Occupational
			0.5	Special Groups
			1.5	Reserve (TV, watches, etc.)
			3.0	
		5.0	(TOTAL)	

* The value 4.5 rem medical dose is the approximate value of the genetically significant dose received in the United States from diagnostic and therapeutic applications of ionizing radiation ("The Genetically Significant Radiation Dose Received by the Population of the United States," a report prepared for the National Academy of Sciences in 1957 by J. S. Laughlin and I. Pullman). The value of 4.5 rem for background is probably an upper limit for most parts of the United States. The sum of the figures 4.5 rem for medical exposure and 5 rem for other exposure is less than the total of 10 rem suggested by a number of geneticists ("The Biological Effects of Atomic Radiation," National Academy of Sciences - National Research Council, 1956) as an upper limit for exposure to man-made sources of ionizing radiation. The 5 rem for other exposure and the breakdown of this 5 rem is suggested by the ICRP ("Recommendations of the International Commission on Radiological Protection," Pergamon Press, London, 1958) only to serve as a guide for planning purposes.

Representative HOLIFIELD. Dr. Morgan, your formal statement will follow your testimony at this point.

(Dr. Morgan's statement follows:)

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SUMMARY AND EVALUATION OF ENVIRONMENTAL FACTORS THAT MUST BE CONSIDERED IN THE DISPOSAL OF RADIOACTIVE WASTE

By

K. Z. Morgan

We have seen from the foregoing discussions that there has been a rapid increase in the amount of radioactive waste; an increase that parallels the growth and expansion of the nuclear energy industry and its associated operations. Every atom of uranium or plutonium that undergoes fission produces, in addition to its 200 Mev of energy, two or more new radioactive atoms. About 3% of these radioactive atoms consist of the more dangerous long-lived strontium-90 (half-life 28 yrs) and cesium-137 (half-life 30 yrs), and traces of the even more dangerous transuranic atoms - americium-241, curium-242, plutonium-239, etc. - are produced. If we assume that in the year 1990 there will be in the U. S. a nuclear power production of 300,000 (3×10^5) Megawatts of heat (which with an electrical conversion efficiency of 25% would be about 10% of the present electrical power production in the U. S.), there will exist in the world one hundred and fifty billion (1.5×10^{11}) curies of radioisotopes from this source including 2.6 billion (2.6×10^9) curies of strontium-90 and 2.3 billion (2.3×10^9) curies of cesium-137. To this must be added the radioactive contamination contributed by nuclear power sources in other countries and the radioactive waste that may be produced by the detonating of nuclear weapons.

It has been indicated that the high level liquid waste may be stored in underground tanks, but this must be considered as a temporary or interim expediency for the integrity of tanks can be relied upon for only a few score

years. It was shown that high level waste may be concentrated so that it can be stored permanently in isolated areas or perhaps much of it can be fabricated into large industrial sources of ionizing radiation to be used as a substitute for X-rays for medical treatment, for the preservation of foods, for mutation experiments in which new farm crops are developed by the irradiation of seeds and plants, for the production of plastics and new chemical products, and perhaps eventually the small-canned radiation sources may become the principal energy units for the operation of equipment in space explorations. Regardless of the many useful applications of these radiation sources, their ultimate disposal presents a problem and the more widespread these useful applications the more universal and ever-present the associated radiation accidents and hazards. We have seen that research is underway in the AEC-supported laboratories to explore the feasibility of various methods of permanent and semi-permanent disposal of high level radioactive waste. Some of these methods consist of fixation in sintered clays and sands, disposal in salt formations, and injections into deep wells, deep sea disposal, etc. These research studies should be continued and the results evaluated from time to time in terms of economics and the long-range radiation protection they can provide.

Radioactive waste is produced in the liquid, gaseous, and solid forms; of these, the liquid waste constitutes the principal long-range environmental problem. This is true, for regardless of the initial physical form, that waste which constitutes a hazard to man is that waste which for the most part has found its way into surface and underground waters. For example, solid radioactive waste is buried in carefully selected and permanently supervised "hot" burial

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grounds or encased in concrete and dropped into ocean deeps. In either case, no radiation hazard ensues unless and until the radioactive waste is leached and carried away from its resting place by dilution or suspension in the water.

Elaborate precautions are taken in the use of filters, precipitators, air washers, and tall stacks to minimize the air contamination problems associated with the nuclear energy industry. Most of the particulate radioactive waste is retained, and only the radioactive Noble gases are discharged in large quantity to the atmosphere through tall stacks. These gases do not present any serious problems, since the maximum permissible concentration (MPC) values of Noble gases are very large and the discharge of the gases at high velocity through tall stacks provides dilution which reduces the concentration at a short distance from the point of discharge far below the MPC. In some operations radioiodine (and in particular iodine-131 with a half-life of 8 days) is discharged from the stacks, and except for accidents (such as the one at Windscale, England, in 1957) this iodine is discharged only into isolated desert areas where it provides valuable information on the uptake by sage brush and prairie grasses and the concentration in the thyroids of wild animals. However, the levels in areas occupied by man are below the MPC values. Most of the environmental radiation problems associated with airborne contamination - regardless of whether they originate from a nuclear power plant or from the testing of weapons - result when the fission products enter the water supply and reach man directly in his drinking water or indirectly by way of vegetation which extracts the fission products from the ground water, or from milk from cattle that graze upon this vegetation.

Unless there are major nuclear accidents it is not the high level waste that introduces most of the environmental radiation problems, but rather the immense volumes of intermediate and low level radioactive waste having a concentration of a few micromicrocuries per cc. Millions of gallons of such waste are discharged daily from a large chemical processing plant. Some of this originates as high level waste from storage tanks or leachings from "hot" burial grounds becoming diluted by large volumes of water in the environment.

Time does not permit a detailed discussion of the values of maximum permissible concentration and how they are obtained, but suffice it to say that in the United States the values of maximum permissible exposure to ionizing radiation are established by the recommendations of the National Committee on Radiation Protection, NCRP[1]. A similar organization, the International Commission on Radiological Protection, ICRP[2], sets these values for international use. These two organizations work closely together, for many of their problems, such as the disposal of radioactive waste, may be of concern to people in many countries of the world. The members of NCRP and ICRP are scientists, i.e., medical men, physicists, biologists, chemists, sanitary engineers, etc., who have specialized in health physics (or radiation protection problems),and they work together as individual scientists - not as representatives of companies, hospitals, laboratories or governments.

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[1] The NCRP was formed in 1929. For information on the history and membership of NCRP see "Brief History of the National Committee on Radiation Protection and Measurements (NCRP) Covering the Period 1929-1946," L. S. Taylor, Health Physics Journal 1: 3-10 (March 1958).

[2] The ICRP was formed in 1926. For information on the history and membership of the ICRP see "History of the International Commission on Radiological Protection (ICRP)," by L. S. Taylor, Health Physics Journal 2: 97-104 (June 1958).

The levels of maximum permissible exposure to ionizing radiation as established by the NCRP and the ICRP are of two types: (1) exposure to external sources, e.g., X-rays from medical equipment, neutrons from high voltage accelerators and nuclear reactors, etc., and (2) exposure to internal sources[3], i.e., radioisotopes taken into the body by inhalation, ingestion, through the pores of the skin, or by way of contaminated wounds.

In considering the environmental factors associated with the disposal of radioactive waste, we are concerned primarily with internal exposure rather than with external exposure. Radioactive material that is taken into the body may deliver its dose in one or in several different ways. If the radioactive material is relatively insoluble, it is likely that most of the dose will be delivered to the lungs or to the gastrointestinal tract. Should it enter the body by way of a puncture wound, most of the dose may be delivered to the small volume of tissue in the vicinity of the wound, resulting in a large localized dose. If the material is very soluble, a large fraction will enter the blood stream. For example, following the ingestion of iodine-131 in milk or water, a measureable amount may enter the blood in a few minutes and 2 to 3% may appear in the thyroid within several hours. In the case of the ingestion of the biologically hazardous, transuranic fissile material, plutonium 239, it is fortunate that only about 0.003% enters the blood. Having entered the blood, portions of

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[3] K. Z. Morgan is Chairman of the Internal Dose Committees of NCRP and ICRP. The Internal Dose handbooks of the NCRP and ICRP have been in the process of revision and were submitted for publication in January 1959. (The NCRP publication will appear as one of the NBS Handbooks available from the Superintendent of Documents, Washington 25, D. C., and the ICRP Handbook will be published by Pergamon Press, London).

the radioactive material are deposited in the various organs of the body. The organ of greatest deposition is usually the critical body organ which is defined as the organ receiving the radioisotope that results in the greatest body damage. Two types of elimination take place simultaneously - biological removal and radioactive decay. For convenience in mathematical calculations, we speak of the effective half-life or the time required for the removal of half of the radioactive material from a body organ. There is great variation in the effective half-life of the radioisotopes. For example, it is 17 years for strontium-90 in the bone and only 37 minutes for chlorine-38 in the total body.

Other important factors that are considered by the NCRP and ICRP in establishing the maximum permissible concentration of the radioisotopes in air, water, and food are mass of the critical body organ, distribution of the isotopes within the critical body organ, type of radiation emitted (e.g., α, β, γ, etc.) energy delivered to the tissue by each disintegration of the radioactive material, curies per gram of radioactive material, etc. With all these factors, it is not surprising that there is a wide variation in the MPC values. For example, the MPC value for occupational exposure (40 hours per week) to tritium is one ten millionth (10^{-7}) curies per cc or 10 grams of tritium per million tons of water, whereas the MPC value for occupational exposure (40 hours per week) to strontium-90 is four million millionths (4×10^{-12}) curies per cc or 0.03 grams of strontium-90 per million tons of water. (For comparison, there are about 6 tons of gold per million tons of sea water.)

There are many uncertainties in the MPC values given in the Internal Dose Handbooks of the NCRP[4] and of the ICRP[5], because in only a few cases are human

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[4] NBS Handbook-52, "Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water." For sale by the Superintendent of Documents, Washington 25, D. C.

5 (see next page)

data available and in some cases even animal data are lacking. However, the latest editions of these handbooks on Internal Dose[3] contain MPC values for about 240 radioisotopes, and these data, although limited in accuracy, represent the best data available at the present time. These data will be revised from time-to-time as more information becomes available. The MPC values given in these handbooks are for occupational exposure. It is expected that in the near future similar MPC values can be recommended for non-occupational exposure. For the present, interim MPC values for persons living in the neighborhood of the nuclear energy operation are to be taken as 10% of the continuous occupational exposure values, and the MPC values for the population at large are to be of the order of natural background (or about 1% of the continuous occupational exposure values when they are based on exposure to the gonads or total body and about 3% of these values when they are based on exposure to other body organs). Because these large reductions in the MPC values are applied to the average exposure to the population at large, the radiation hazards of the nuclear energy industry are restricted primarily to occupational workers. This is in conformance with the usual practice of restricting the unique hazards of each occupation to the workers in the occupational group whether it be bridge building, housekeeping, or nuclear energy. Thanks to the fine job done by the health physicists, the average exposure of the occupational worker in the nuclear energy industry is only a few percent of the maximum permissible value and other hazards have been

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[5] "Recommendations of the International Commission on Radiological Protection," British Journal of Radiology, Supplement No. 6 (1955).

reduced correspondingly so that the nuclear energy industry in the United States has maintained one of the best records of safety.

As explained above, the MPC values presently recommended and published by the NCRP and the ICRP were selected primarily for application to the occupational worker. At best the present ad interim application to the population at large can serve only as a guide until we obtain more information on the genetic and ecological effects of radiation on man and his environment. The arduous task of developing tables of MPC values that are specifically applicable to the population at large has already begun, and it is anticipated that some of the data will be available for publication in about three years. It is evident that three types of damage must be considered: (1) somatic damage or direct injury to the individual, e.g., the induction of leukemia and shortening of life span, (2) genetic damage appearing in those to be born, and (3) indirect damage to man's environment, e.g., the effect on his food supply and the natural resources. These first two effects have been mentioned by the other speakers and discussed in some detail in the hearings on Fallout[6] and in reports of the United Nations Scientific Committee[7]. Therefore, in what follows we will limit our discussion to the ecological effects of radiation on the environment.

There seems to be a rather widespread feeling, especially among laboratory biologists, that since man seems to be as radiosensitive as any organism all

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6 "The Nature of Radioactive Fallout and Its Effects on Man," Hearings before the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy Congress of the United States, Part I (May 27, 28, and 29 and June 3, 1957); Part II (June 4, 5, 6, and 7, 1957), Government Printing Office, Washington, D. C.

7 "Report of the United Nations Scientific Committee on the Effects of Atomic Radiation," General Assembly Official Records: Thirteenth Session, Supplement No. 17 (A/3838), United Nations, New York (1958).

we need do is "monitor" radiation in the environment where man actually lives. One authority[8] has stated it thus: "It is our belief that, if we take sufficient care radio-biologically to look after mankind, with few exceptions the rest of nature will take care of itself. Sensitivity to ionizing radiations is well correlated with degree of evolutionary complexity. Man and his domestic mammals will probably be among the first to suffer from overexposure to radiation." Perhaps this is a dangerous oversimplification.

In the first place, there are many important environments such as lakes, streams, and soil where man himself is not a direct resident but upon which he depends indirectly for his food, building materials, pleasure, etc. The possible effects on man resulting from contamination of these environments by radioactive materials cannot be evaluated without a study of these environments themselves. Second, these environments contain populations of plants and animals which are organized into biological systems or communities, and man, insofar as we know now, cannot survive independently. Communities and members of communities are dependent upon each other for their continued existence, and this type of biological organization with its complex cycles of elements, compounds, and minerals is characteristic of our entire world. Therefore, in any waste disposal program we must take into consideration not only the problem of maintaining concentrations of radioactive material in soil and water at levels considered to be safe for direct exposure to man and his children (i.e., safe from the standpoint of somatic and genetic damage), but also we must consider the possible consequences of the escape and accumulation of small quantities of radioactive

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[8] Loutit, John F., "The Experimental Animal for Study of the Biological Effects of Radiation," Peaceful Uses of Atomic Energy, Vol. XI, p. 3 (United Nations, New York 1956).

materials into the environment where long-range modifications may result in a less favorable place in which to live. In a large-scale nuclear power economy where there is competition for cheaper sources of energy, there will be an increasing need and demand to make use of the dilutent capacity of the environment for the economic disposal of large volumes of low level radioactive wastes. Thus, long-range continuing studies must be made to assure that the levels of radioactive contamination in the environment are not kept so low as to unnecessarily impede this new industry or, on the other hand, are permitted to become so large that the damage to the environment exceeds the over-all benefits from nuclear power and its useful products. What concerns us here is not the massive effects that might result from the accidental release of large quantities of fission products, but rather the subtle cumulative effects that might be building up due to the slow but continuously increasing radioactive contamination of our environment. Such effects may not become manifest or may remain concealed due to other more pronounced phenomena and may only become evident hundreds of years from now. Because of this uncertainty in our knowledge, we believe that environmental research programs should be undertaken and should be specifically oriented toward the contamination problem. These programs should not only be corollaries of our waste disposal research efforts which are mainly concerned with safe methods of disposal, but also these environmental programs should provide us with the necessary basic and applied information so that undesirable trends may be recognized in time for remedial action to be taken before unsafe conditions begin to appear.

I should like to point out that there are two phases to this problem of environmental contamination and its possible effects on man. The first is the effect of continuous low level radiation on ecological systems and on the complex

relationships between animals, plants, arthropods, bacteria, and fungi. The second is the ultimate fate of radionuclides in these natural systems. Actually these two phases are closely interrelated, because the radionuclide is the source of radiation in the system, and the chemical and biological transfer of the radionuclides within such systems will determine the extent to which radiation will be present at the various levels of organization in these systems.

There are many examples of instances in which man has accidentally or intentionally tampered with his environments and produced unexpected results. Early work in wildlife management and the introduction of animals into new areas often produced the opposite effect from the desired one. More recently, the widespread use of the synthetic insecticides has created new insect problems as well as solved old ones.

Almost nothing is known about the possible effects of low level long-continued exposure to man-made sources of radiation. But we can at least approach the subject by examining the considerable body of data on radiosensitivity. Even before the current upsurge of interest in the biological effects of ionizing radiations, enough work had been done with X-rays to indicate that organisms differed widely in their ability to tolerate heavy doses. Recent work has served to show that the subject of comparative radiosensitivity is extremely complex. It should be emphasized that because of experimental limitations doses used in most studies have been much higher than we expect to have to contend with in the general environment. Nevertheless, there is evidence that has bearing on the low level chronic effect. Suppose we make the simplifying assumption that because man is more sensitive to an acute dose (i.e., a large, single dose) than are bacteria, that he is more likely to be affected by low level chronic doses. Based on the data now available, and if this assumption

were valid, we would expect mammals to be the most sensitive and bacteria among the least sensitive of organisms. Plants, lower vertebrates, insects, and other invertebrates would fall somewhere between these extremes. But there is a flaw or oversimplification in this argument from the ecological point of view. The assumption cannot be upheld since organisms do not have a constant radiosensitivity throughout their life span. Even the most resistant groups have stages in their life cycles or critical periods in their population growth which are affected by much lower doses of radiation. In the fruit fly, Drosophila, which is one of the more resistant insects, a dose in the neighborhood of 100,000 roentgens is required to kill half of the adult flies (mid-lethal dose) while as little as 160 roentgens may kill half the individuals at the most sensitive stage in the fly's development (which is the egg during cleavage). In other words, at this egg stage the susceptibility of the organism is over 600 times that of the adult stage. A dose as low as 8 roentgens per day has been shown to have a measurable effect on the growth and development of grasshopper nerve cells. Likewise, barley, which in common with most seed plants is relatively radio-resistant as a mature plant, is measurably affected during the early stages of development by doses no higher than that which would be critical for mammals. Consequently, when we consider the vulnerable stages, we find that some of the wide differences which exist between adults of different taxonomic groups have disappeared.

When radioactive isotopes are released into the environment, they usually enter into complex physical and biological cycles and into food chains and may exhibit unexpected movements or concentrations. It is through such mechanisms that dangerous long-lived radionuclides such as strontium-90 and cesium-137 are able to enter our food sources. These ecological food chains are actually

circular in nature or web-like and have a limited number of links. Serious consequences could result if too many of these food links were broken. Radionuclides with long half-lives may be expected to be taken up and dispersed, and reconcentrated by ecological factors, although no one has yet followed materials through more than one cycle. The rates at which these cycles operate constitute one of the most important and least-known aspects of ecology and have important bearing upon the release of radioactive wastes into any environment.

Evidence of selective absorption factors for some radioisotopes as high as 100,000 has been obtained for certain organisms. Soil concentrations of fission products of one billionth of a curie per gram are common in the Oak Ridge waste disposal areas. If the organism is in equilibrium with its immediate environment it can have dose rates ranging from 10 to 1,000,000 rads per year depending on whether there was a selective absorption factor of one or of 100,000. At this point it should be recalled that only a few organisms have been examined with respect to their radiosensitivity and almost none for the effect of continuous exposure to radiation over a long period of time. In particular we should note that studies have been made with respect to killing effect; and that there may be many other types of damage more subtle but just as damaging in consequence to the human environment as immediate death. For example, radiation may affect the mobility of the organisms, or the availability of kinetic energy, such that they are unable to obtain a sufficient amount of food or engage in other useful functions. It may affect the germ cells such that there is a reduction in fertility. It may affect visibility or other sensory functions such as to incapacitate the organism. Also, if a small dose has any effect at all at any stage in the life cycle of an organism, then chronic radiation at this level can be more damaging than a single massive dose which comes at a time when only

resistant stages are present. Finally, stress and changes in mutation rates may be produced even when there is no immediately obvious effect on survival of irradiated individuals.

A person might object to the use of a factor of selective absorption of 100,000. This, or perhaps even higher factors, may obtain for certain links in the complicated ecological web. Or the objection may be raised that perhaps these links can be wiped out and the ecological universe will have the capacity to readjust itself and reach a new type of equilibrium that will satisfactorily meet the requirements of man and his environment. I do not believe these are valid objections, at least for the health physicist, until they are supported by experimental evidence. To this end we have encouraged the development of ecological research programs which are based on, and are integral parts of, radioactive waste disposal research programs at Oak Ridge National Laboratory.

Conclusions

1. Radioactive contamination in the environment can be expected to increase in proportion to increases in the nuclear power industry.
2. It is not expected that the present interim radiation exposure levels for the population at large will be exceeded in the foreseeable future.
3. At present, medical exposure to ionizing radiation constitutes essentially all of man's radiation exposure (See Table 1). In the U. S.,medical exposure probably is slightly greater than man's exposure to the natural background radiation[7].
4. If the present level of average medical exposure is maintained in the U. S. and if the interim MPC levels for the population at large are reached (say within 20 to 50 years), the radioactive waste will contribute an exposure to man that is equal to only 1/3 the exposure received from medical sources. (See table 1.)

Table 1, referred to above appears on p. 2377.

5. The somatic and genetic consequences or damage from this exposure depends upon the existence (or lack) of a linear relationship between radiation dose and effect at these very low dose rates. This subject has been discussed elsewhere and, therefore, is not repeated here. In any case if in the future the present MPC levels are reached, waste disposal will contribute no more than 1/3 the somatic and genetic damage received by man at the present time from medical sources.

6. The effect of exposure of radioactive waste on the environment is unknown. The average dose delivered to the environment at the present time by man-made radioactive waste is very low, and even in the future if the presently recommended levels of MPC are reached, the average dose probably will be no more than 50% of that from natural background. However, because of selective deposition certain elements in the food web may concentrate some of the long-lived radionuclides, and consequently critical links in the food web might receive local doses that are very high. The end result could be a reduction in man's food supply or a destruction of some of the natural resources.

7. Many AEC-supported programs are in progress to determine safe and relatively permanent methods of radioactive waste disposal; to evaluate the somatic, genetic, and environmental damage produced by chronic exposure to low level radioactive waste products; and to establish reliable MPC values for each of the radionuclides. This is a monumental task, and many interesting research data have been obtained already, but much remains to be done requiring research studies into the distant future.

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8. Although radiation hazards of new and heretofore undreamed of proportions have arisen in the nuclear energy industry, this work has been done safely. With the untiring efforts and constant vigilance of the health physicists, this has become one of the safest of American industries. With continued alertness and with an increased number of long-range health physics research programs, this record can be maintained and the benefits of nuclear energy and the useful application of the radioactive products can be made to dwarf by comparison any harmful effects of radiation on man and his environment.

Representative HOLIFIELD. Our next witness will be Mr. J. G. Terrill of the U.S. Public Health Service.

STATEMENT OF JAMES G. TERRILL, JR.,[4] ASSISTANT CHIEF, DIVISION OF RADIOLOGICAL HEALTH, U.S. PUBLIC HEALTH SERVICE

Mr. TERRILL. Mr. Chairman, and members of the committee, I would like to summarize my speech in the interest of time, but I would wish to ask that the original copy be inserted in the record.

Representative HOLIFIELD. It will be received at this time.

(The statement is as follows:)

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[4] Graduated from the University of Cincinnati in 1937 with a degree in civil engineering. He studied public health engineering at the Massachusetts Institute of Technology Graduate School from 1938–41. Since 1941 he has been active in the Public Health Service. He participated in the first Bikini tests. During the period 1948–51 he studied radiological defense under the sponsorship of the Armed Forces Special Weapons project at the U.S. Navy Postgraduate School and the University of California. He has participated in and directed the Public Health Service activities related to the Nevada and Pacific test operations during 1953–57.

Mr. Terrill is active in radiological committees of the American Society of Civil Engineers and the American Public Health Association. He is a member of the National Committee on Radiation Protection and the Nuclear Standards Board of the American Standards Association.

Presently, he is Assistant Chief of the Division of Radiological Health, Public Health Service, with headquarters at 4th and Independence Avenue SW., Washington, D.C. Mr. Terrill lives at 5812 Phoenix Drive, Bethesda 14, Md.

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Future Estimates and Economics of Waste Disposal Summary and Evaluation of Environmental Factors

By

James G. Terrill, Jr., Assistant Chief, Division of Radiological Health, U.S. Public Health Service.

Introduction

In view of the comprehensive nature of these hearings and the excellent presentations made by the other speakers, my remarks will be confined to those aspects of radioactive waste disposal activities that our experience in the Public Health Service indicates to be of major importance from a public health standpoint and through this, comment on economics of waste disposal principally as it may affect health agencies. Dr. Price* Chief of the Bureau of State Services, will discuss activities of the Public Health Service relating to radioactive wastes tomorrow, and Dr. Ernest Tsivoglou** has presented a paper for the record on surface waters. The experiences that are about to be presented, while expressing a Public Health Service viewpoint, are related to the activities reported by many of the other speakers and a continuing cooperative effort with many of the divisions of the Atomic Energy Commission, but particularly with the Division of Reactor Development.

General Health Principles

From the public health standpoint the primary consideration is the overall effect on people of exposure to all types of radiation in the expected but variable pattern of sickness, health, and age. Consequently, even though we are principally dealing for purposes of these hearings, with the question of disposal of radioactive wastes, we must bear in mind that this is only one

* Dr. David E. Price, Chief, Bureau of State Services, PHS

** Dr. Ernest C. Tsivoglou, Senior Sanitary Engineer, Water Supply and Water Pollution, Robert A. Taft Sanitary Engineering Center, PHS.

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facet of possible radiation exposure that must be considered by health agencies. In addition to external exposures from X-ray and background, human populations may ingest or inhale radioactive materials. Therefore, in determining criteria for the safe disposal of radioactive wastes all means of exposure of local or national population must be considered. In view of these principles it is most important to know the current concentration of radioactive materials in air, foods, and drinking water, the existing protection mechanisms, and the basis for current radiation standards if they exist.

Water Decontamination Studies

With these thoughts in mind, soon after the creation of the Atomic Energy Commission, the Public Health Service developed a research program cooperatively with the Commission and the Department of Defense, and with the advice of several professional organizations to learn quantitatively and qualitatively about radioactive wastes and the effectiveness of existing water treatment facilities when applied to raw waters contaminated by radioactive materials. The results of the early stream studies have been presented by Dr. Tsivoglou in his reference to the Columbia River studies (1) and the Savannah River studies (2). Concurrently the experimental work on decontamination of radioactive waters was undertaken at Oak Ridge. The results of these studies may be summarized in the following way. (3)

In the water treatment processes involving coagulation, settling and filtration, most of the radioactive particles will be removed, but only about 10 - 70% of the dissolved radionuclides will be removed. Ion exchange resins can be used to reduce the dissolved material by 99%. Another effective but costly process is distillation. Neither of these processes, however, is sufficiently economical to encourage their use as a method which could be utilized by large cities and industries to protect themselves from abnormally high concentrations of radioactivity.

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It was our judgment after a major portion of these studies were completed, that first consideration should be given to the control of radioactive wastes at their source, and secondly to alternative water supplies, preferably from ground water sources which are known to have low concentrations of natural radioactivity. This tentative conclusion was strengthened by continuing research carried out by the Public Health Service as a part of the basic program, and for the Department of the Navy, the Air Force, and the Atomic Energy Commission. (4)

Environmental measurements undertaken as part of the radiological surveillance activities of the Public Health Service (an example of which is contained in Appendix A) indicated that the water works operator and the health official were not only confronted with natural radioactivity and possible waste materials from reactors and radioisotopes, but that very appreciable quantities of gross radioactivity were being deposited on the ground in the rainfall and in our water courses from the nuclear debris from atomic weapons tests. Concurrently, however, our continuing liaison with major installations handling industrial atomic eastes, such as Oak Ridge, the Savannah River plant, and the Idaho Operations Office, indicated that these installations were sufficiently isolated and controlled so the major producers of environmental activity at these installations probably would not be a cause of public health concern in the near future.

This background material defines at least qualitatively the impact of radioactive wastes and associated activity on both the industry and the public agencies responsible for the safety of water supplies. It appears that the atomic waste treatment facilities should be asked to produce an effluent of sufficient purity so the water at the intakes of water treatment plants will be as close to drinking water standards as general environmental contamination will allow.

If the raw waters are contaminated, the waterworks operator and the health agencies which assure the safety of the water supplies, should first initiate continuing measurement and evaluation of radiological quality. If these relatively simple technics fail to assure a safe water supply radiologically, those responsible are confronted with major changes in their basic water supply planning either from the standpoint of sources or treatment or both. Estimates of the cost of such major changes in water supply systems even on a single river system would be a major engineering effort.

Thus the economics of waste disposal involves both the industrial unit producing the waste, and the agencies responsible for the safety of public water supplies.

Wastes at Power Reactor Sites

With the foregoing considerations in mind, a few years ago the limited resources of the Public Health Service were shifted to activities which would have the most immediate impact on radiation emanating from power reactors, and the measurement and evaluation of all of the radioactivity which is found in the environment.

An opportunity for both study, service, and experience became available during the planning of the Shippingport reactor. It was our belief that this reactor system could serve as a prototype for the development of a satisfactory technical and administrative pre- and post-operational evaluation system of a typical power reactor. Through the cooperation of the AEC Naval Reactors Branch, a quantitative report on the prospective wastes, an evaluation of the waste treatment system, and its probable effects on the river were developed. (5 and 6) With the State of Pennsylvania and the Pittsburgh Operations Office of AEC an administrative instrument in the form of a waste disposal permit was developed which was approved by the State Board of Health to serve as the basis of both administrative action and public understanding (Appendix B). The Public Health Service has continued its

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assignment of personnel to the Pittsburgh Operations Office, and that agency has continued to supply data which illustrate the type of information which public health agencies must tabulate and analyze to assure against over-exposure. (Appendix C)

It now appears that the prospective operator of nuclear reactors is confronted with a complex environmental situation in which the radioactivity being discharged to the environment is mixed in with other natural and man-made radioactivity in similar concentrations. While these concentrations are low, they are in a range which fluctuates around a level of activity of 10^{-8} uc/cc, which has been considered to be the level at which further identification of radionuclides is necessary to assure public exposure below maximum permissible levels.

High Level Wastes

The Public Health Service has made estimates of the potentialities of high level waste production (7) and has tried to inform the health departments through its training courses and writings that the majority of the fission products which are produced would decay within the reactor system or be handled in chemical processing plants. Our calculations for given power levels generally parallel those made by the earlier speakers, such as Dr. Culler in his published work. The principal variable in these calculations is the predicted nuclear power level. (8)

For several years, the Public Health Service worked with the AEC at Oak Ridge on the disposal of intermediate wastes. Currently our principal association with relatively high level material is through our cooperative project at the Idaho Operations Office. This involves environmental measurements of the emanations from both reactors and from the chemical processing facility there. In addition, one of our trainees at the Massachusetts Institute of Technology, working on an AEC project with Professor Eliassen, has developed a ceramic material which may be useful in some types of relatively

high level waste disposal (9).

The principal emphasis of the AEC high level waste research is reported as the selective disposal of hazardous isotopes, ocean disposal, subterranean disposal in salt domes, ion exchange in soils, fixation in ceramic materials, pyro processing, and economic utilization of fission products. In practically every case the development of a feasible method for handling of these wastes involves questions of health either during transportation, chemical processing, or disposal. (8) Therefore, health agencies are vitally interested in this research particularly regarding the relative health hazards involved in each system of transportation, chemical processing and ultimate disposal.

Operationally, the health agencies understand that tank storage is the present method of choice for high level wastes. These storage facilities are now located at a relatively few government-operated processing plants. In our planning it is expected that this situation will continue and that the economic burden of this phase of the disposal problem will largely rest on the Federal Government during the immediate future.

Limited staff work indicates that the cost estimates of high level waste disposal in future years, reported by Culler and others, are quite reasonable, and that the Nation can expect that the operational requirements, insurance, and radiation protection requirements related to the handling, transportation, processing and disposal of high level radioactive wastes will require multimillion dollar expenditures. The portion of this expenditure, which will be placed upon health agencies, local communities, and individual industries, will depend upon many factors including acceptable health standards and the extent to which major changes in nuclear fueling and reactor operations develop, as well as factors related directly to waste handling and disposal.

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Application of Radioactive Material

Conventional, Industrial and Medical Uses

The Atomic Energy Commission has reported the distribution of several hundred thousand curies of radioactivity to qualified purchasers in over a hundred thousand shipments. These are principally for medical, industrial, and agricultural research purposes. However, of this amount, a substantial portion is in the form of sealed sources of radioactive cobalt-60 and iridium. These sealed sources do not normally constitute a potential environmental contamination hazard in the usual sense. Radioactive iodine and phosphorus are other isotopes which might reach the air and water in appreciable quantities. Both iodine-131 and phosphorus-32 have short half-lives which automatically limit their contribution to exposure. Thus, in a comparison with the waste potentialities of fuel processing plants and reactors, it would appear that the environmental hazards from the isotopes program are associated primarily with their widespread distribution, transportation, and variability of the technical competence of the users, rather than with their magnitude or the relative hazard of the specific isotopes involved. From the standpoint of radiological public health costs, it would appear that the requirements for personnel and equipment of this phase of waste disposal would not place an identifiable burden upon health and related agencies which in the future should provide themselves with appropriate competencies for their other activities applicable to the use and disposal of radioactive materials.

Environmental Tracers and Peaceful Uses of Nuclear Detonations

Tracer studies using large quantities of long-lived activity such as tritium and the peaceful uses of nuclear detonations for industrial applications have been proposed recently. These applications of nuclear energy have not been studied in detail by the Public Health Service. At this time we would like to clearly differentiate between the hazard potentialities of these applications and those of conventional industrial and medical uses.

Some of the proposals may require a clearer delineation of the benefits to be derived, administrative and financial responsibilities, correlation with other radiation hazards in the area, and a review by a variety of scientific competencies and public agencies, in addition to normal health physics considerations.

Costs of the associated radiation safety activities which the public health agencies may be expected to bear have not been estimated by the Public Health Service. Our experience with health and safety operations related to reactors and weapons testing suggest that considerable expenditures will be required by the State and local agencies to carry out their responsibilities.

Preoperational and Operational Surveys

The experiences described in the previous sections have often led to planning and executing minimum radiological surveys in cooperation with many Federal and State agencies. Nearly all of these involve cooperation with the AEC. They include the Navy shipyards; the NACA reactor, and the State of Ohio; the Elk River reactor and the State of Minnesota, the Camden-Philadelphia area with regard to the N. S. SAVANNAH; and the State of Michigan relative to the reactor near Monroe. The more complete studies of the Ohio, Columbia, Savannah and Animas Rivers have been reported by Dr. Tsivoglou.

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An understanding of the reactor type and details of its waste handling plans is necessary in every instance to plan for the sampling and analysis in a significant way. The execution surveys even on a minimal basis will place further fiscal requirements on the government and industries concerned as the nuclear industry develops. The costs will also be affected by existing levels of radiation in the environment which often preclude reliance upon gross counting technics. (10 and 11)

Generally, we have encouraged reactor planners and health agencies to obtain minimal amounts of pre-operational data in the areas surrounding the reactors both as a check on their safety and waste systems, and also to have facts available when high levels of radioactivity are found so the emanations from the reactor system per se may quickly be identified, and where possible controlled.

Methodology

The cost of measuring radioactivity has led the Public Health Service to intensify its research on methodology. Engineering studies show the presence of radioactive materials in all phases of the environment. Gross determinations, although of value in indicating trends in activity level, are not readily related to hazard. Because of their radiological, biological, chemical, and physical characteristics, each radionuclide has its own maximum permissible concentration in air, water, milk, or food. In applying these values one should not neglect the hazards associated with stream uses over and beyond use of water for domestic purposes. External hazards associated with swimming and the industrial use of water must also be considered. (12, 13)

The task of determining specific radionuclides in environmental media is not simple. Extremely low concentrations of the radionuclides must be separated from very large amounts of stable and inert materials. Thus,

sample preparation for the separation of individual radionuclides presents one of the greatest problems. For example, the determination of an isotope such as strontium-90 in sea water, milk, biological materials, etc., is quite different from its determination in fresh water, soil, or air. Much effort has been expended, therefore, on the part of the Public Health Service, to develop methods for the separation and identification of the more hazardous radionuclides in these environmental media. Essentially, the approach has been to look for the most hazardous radionuclide first. If it is absent, one looks for the next most hazardous, etc., until the major more hazardous radionuclide activities have been identified or shown to be absent. Methods have been published for the identification of total radiostrontium, strontium-90, strontium-89, barium-140, cesium-137, iodine-131, cerium-141, cerium-144, ruthenium-103, ruthenium-106, yttrium-90 and radium-226 in water (12-16); the same nuclides plus cobalt-60 adsorbed on bottom muds (17); modifications of the above with soils; total radiostrontium, strontium-89, strontium-90, barium-140, iodine-131, and cesium-137 in milk (18); and the qualitative determination of many radionuclides in a variety of materials by gamma spectrometry (19-20). Currently, methodology is being developed to separate and identify these radionuclides in estuarine and marine water and associated biota.

With the means for identifying the specific radionuclides at hand, and the variety of environmental samples available through the current surveillance operations of the Public Health Service, some estimates of each of the more hazardous radionuclides affecting man can be estimated. For example, from the intake of strontium-90 through such vehicles as air, water, milk, and other foods, an estimated total strontium-90 body burden can be calculated as a

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function of concentration and period of intake. This can then be related to the dose. In the same manner, the radiation dose contributed by each of the other radionuclides can be estimated. To these would be added the exposure from naturally occurring radioactive materials, the external radiation from cosmic rays and naturally occurring radionuclides, and from x-ray and other ionization-producing devices. Thus, in the future, man's total exposure may be estimated.

The maximum permissible concentrations for radionuclides in any environmental phase relate specifically to the manner in which that phase is employed by man. If the air and water in a particular area contain radioactive materials, it is quite probable that milk and foodstuffs produced in that environment will also contain radioactive materials. Because activity levels in water meet criteria for drinking, or levels in air meet criteria for breathing, does not necessarily mean milk or foodstuffs from the area will also meet acceptable standards. Measurements covering one environmental phase are not adequate for estimating the hazard due to another phase. Monitoring based on such a procedure can lead to a false sense of security since the real hazard is not always the obvious hazard.

The studies now under way involve review and adaptation of technics and data supplied by the AEC, development of ideas originated by the Public Health Service staff, and field testing at Idaho Operations Office and at weapons testing installations. In air monitoring around reactors, a very useful item which has been tested is an activated carbon iodine sampler developed at the Idaho Operations Office. This can be used to collect radioactive iodine in the gaseous form in the environment, and could provide data on the concentration of iodine in air, which conventional particulate samplers miss.

Health Standards

The need for health standards developed through occupational hazards associated with the use of x-ray machines and radium. Later this was supplemented by the more diverse requirements of the military atomic energy developments and by the industrial applications sponsored by the Atomic Energy Commission. As you know, from previous hearings, the levels for non-occupational purposes which are currently being used are adaptations of the levels prescribed for occupational purposes. In recent years many scientific people have raised questions about genetic as well as somatic effects of radiation at low levels over long periods of time. This is the area of particular importance to public health officials. These questions involve the consideration of many factors such as tissue dose, susceptibility of tissues, variation in the distribution of internally deposited substances, the symbiotic effects of multiple exposures, variability of effects with age, the existence or non-existence of threshold phenomena, and other matters which have been considered in detail by your Committee on previous occasions.

However, even though there are differences of opinion on the precise level at which significant health effects may be produced, it seems to be the scientific consensus that radiation levels closely corresponding to those often found in the natural background can serve as operating recommendations at the present time for the more significant isotopes. In the past, the Public Health Service has made every effort to be as consistent with the National Committee on Radiation Protection recommendations as interpretation of those recommendations and methodology would allow. Looking to the future, however, it is apparent that very serious thought must be given to the fundamental factors mentioned above and to details such as the units in which the radioactivity will be expressed, i.e., micromicrocuries per liter

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vis-a-vis micromicrocuries of strontium per gram of calcium, since interpretations, calculations, and laboratory work will be extensively affected by the simple choice of units. Generally, the strontium unit offers many advantages in terms of evaluating milk. In evaluating water affected by radioactivity its use could lead to excessive concern where waters of low calcium content are involved. The terminology of "maximum permissible" while offering many advantages where occupational exposure is involved, presents many difficulties in interpretation of the health significance of the levels of a mixture of fallout and reactor effluent which will be measured by the public health agencies.

It is our hope that the scientific groups developing standards and providing the research data on which standards are based will give consideration to the establishment of several levels at which health agencies should take appropriate action consistent with risk, rather than to rely entirely upon one level and to define it as maximum permissible. The levels that are finally chosen may have more effect on the economics of waste handling and disposal both in the construction and operation stages than any other single factor related to health. Likewise, the requirements of the health agencies for staff and equipment to perform the essential health measurements and evaluations will be affected in a major way by the standards that are chosen.

Summary

It is our current belief that the over-all radiation in the environment in the country will become more complex, and the radiation levels will generally tend to increase to a point where current health standards will be threatened. A controlling factor in the development of the atomic energy industry may be the economics of waste disposal methods necessary to meet

these criteria. Thus, it is of great national importance that a firmer basis for the health standards be developed as rapidly as possible, and that economical and significant methods of analyses which can be related to these standards be developed and approved by appropriate professional organizations. It is expected that the Public Health Service concurrently must apply its resources to the technical assistance, research, and training programs in a major way so that the health agencies can contribute to the peaceful development of atomic energy with a minimum exposure of the people to ionizing radiations.

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APPENDICES

Appendix A (See page 72)	"The Nature of Wastes Produced in the Mining and Milling of Uranium Ores" by Duncan A. Holaday, Occupational Health Field Station, Salt Lake City, Utah, Occupational Health Program, Public Health Service.
Appendix B (See page 2811)	Industrial Wastes Permit #1832, Commonwealth of Pennsylvania, Department of Health, Sanitary Water Board, Harrisburg.
Appendix C	PWR - Waste Operation Experience from Duquesne - PNROO - PHS Liaison - Summarized by Special Projects Branch - Division of Radiological Health - Jan. 12, 1959.
Appendix D	Training Activities, Division of Radiological Health - January 1959.

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APPENDIX C

PWR - WASTE OPERATIONS EXPERIENCE
from
DUQUESNE - PNROO - PHS LIAISON - Summarized by SPECIAL PROJECTS BRANCH
DIVISION OF RADIOLOGICAL HEALTH

The accompanying graphs represent a monthly summary of the volume of liquid, and quantities of radioactive material released to the Ohio River as compared to the design criteria. Attention has been paid only to liquid and gaseous wastes since there is no possibility of solid waste being released to the public environment. Airborne particulate contamination is also effectively eliminated by the filtering and scrubbing of the gases resulting from the incineration of combustible solids. The gaseous activity is then limited to gaseous fission products (Kr and Xo) removed from the reactor plant effluent.

Of the other activity contained within the resultant waste, approximately 20% consists of Tritium with the remaining 80% being due to the fission and corrosion products in the reactor coolant. Of this latter 99.34% will be removed by decay or by ion exchange resins before the wastes are discharged to the river or to the atmosphere; 0.17% will be discharged to the atmosphere; 0.0034% to the Ohio river, and 0.4866% is contained in the wastes which are drummed for ocean disposal.

Neither special waste, consisting of liquid from the "hot" laundries and the laboratory drains, nor the reactor plant effluent, which may be processed, if necessary, through ion exchange resin,

and a gas stripper which removes the Kr and Xo, exceeded the design criteria. Special wastes contributed 6300uc for the 10 month period as compared to a design criteria for 32,200uc for 10 months. In the same period the reactor plant effluent contributed 26,900uc of corrosion and fission product activity as compared with an estimate of 38,400uc, and approximately 46 curies of Tritium versus and estimated 3000 curies. The total gross activity released, exclusive of Tritium, was 34,812uc as compared to 47,700uc per month design. Referring back to the graphs, the gross activity discharge to the river is a summation of 3 types which are shown individually in terms of the exchange activity released per month in uc/ml. Non-active waste consists of wash water from the "cold" laundry and from the showers, twice exceeded the design criteria in the terms of uc/ml within the first 10 months of 1958, however the total activity released in this period 1613uc was well below the estimated value of 6450uc for 10 months.

Release of the stored gases will occur approximately every four months. During 1958, 37,581uc of activity, principally Kr^{85} due to decay of the other fission product gases during the storage period, were released. The design criteria provides for the release of 2,000,000uc per month.

In some cases the volumes of liquid and gases released exceeded the design criteria, but since the total amount of activity was less than design, this only served to further lower the concentrations uc/ml below the design values.

Fig. 1 TOTAL GROSS GAMMA ACTIVITY DISCHARGED TO OHIO RIVER FOR 1958

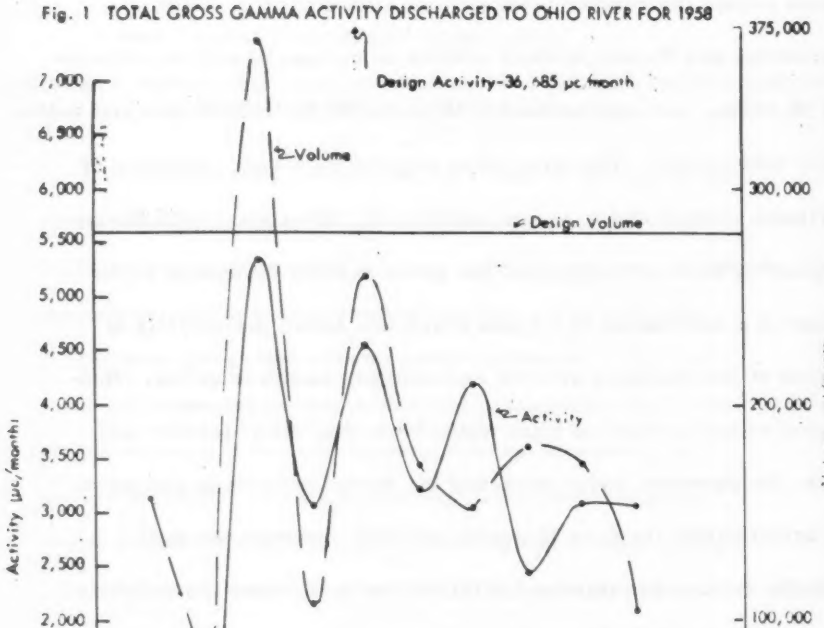


Fig. 2 REACTOR PLANT EFFLUENT DISCHARGED TO OHIO RIVER - 1958, GROSS GAMMA ACTIVITY

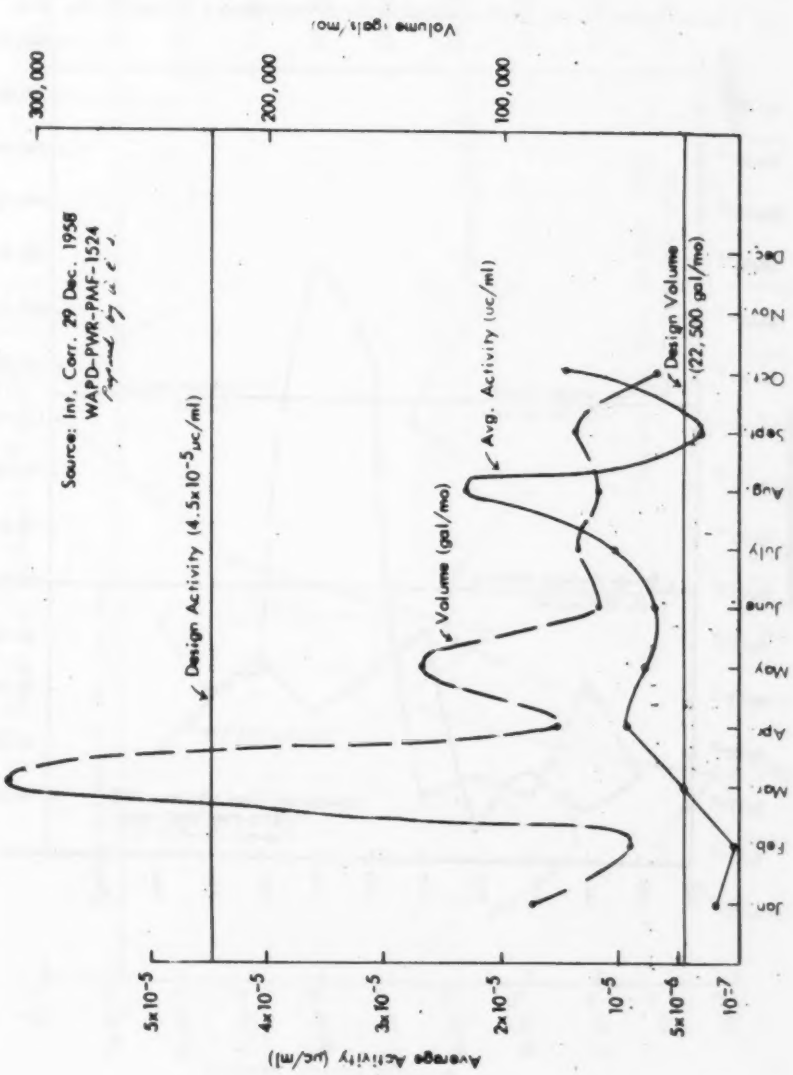


Fig. 2 REACTOR PLANT EFFLUENT DISCHARGED TO OHIO RIVER - 1958, GROSS GAMMA ACTIVITY

Fig. 3 NON-ACTIVE WASTE, GROSS GAMMA ACTIVITY DISCHARGED TO OHIO RIVER, 1958

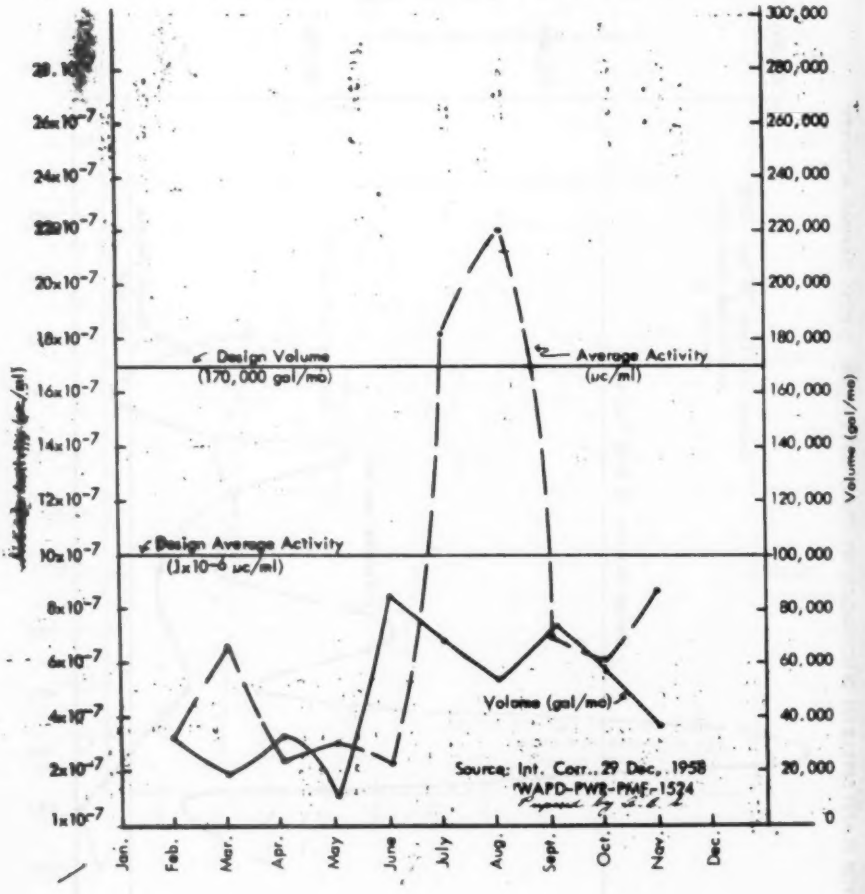


Fig. 4 SPECIAL WASTE DISCHARGED TO OHIO RIVER - 1958, GROSS GAMMA ACTIVITY

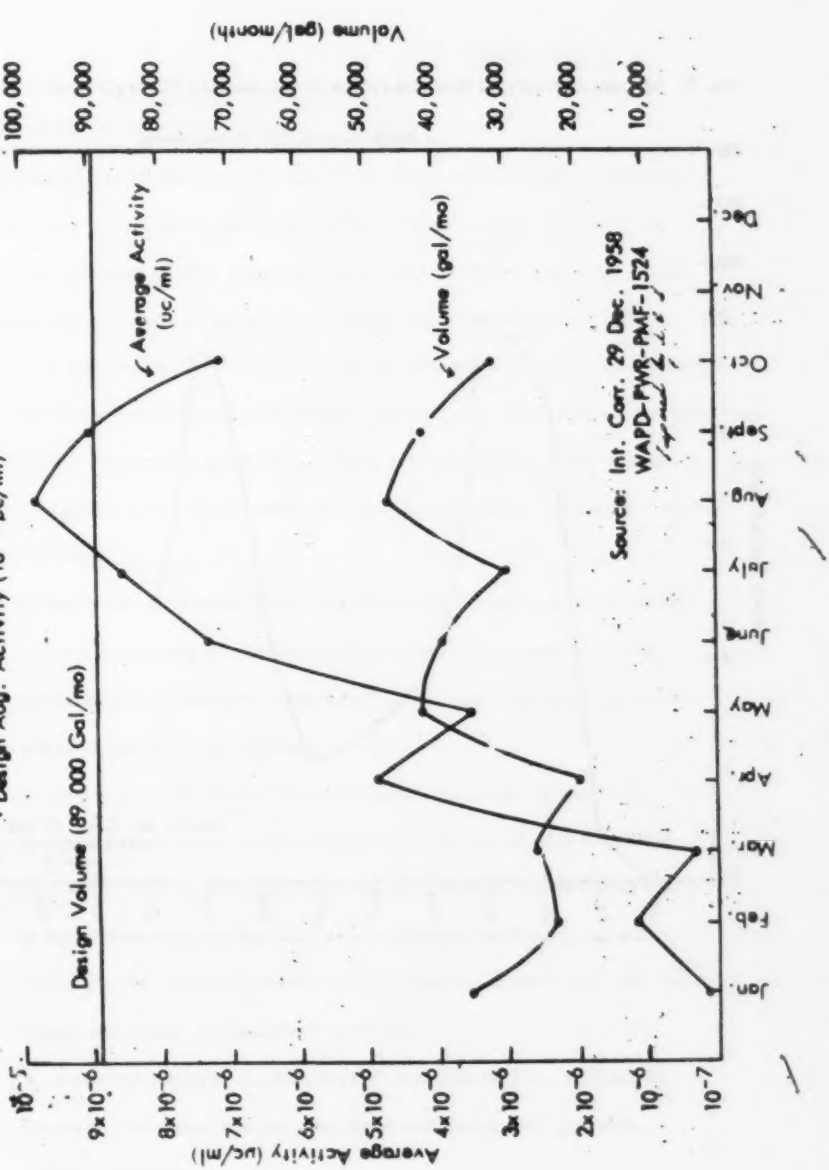
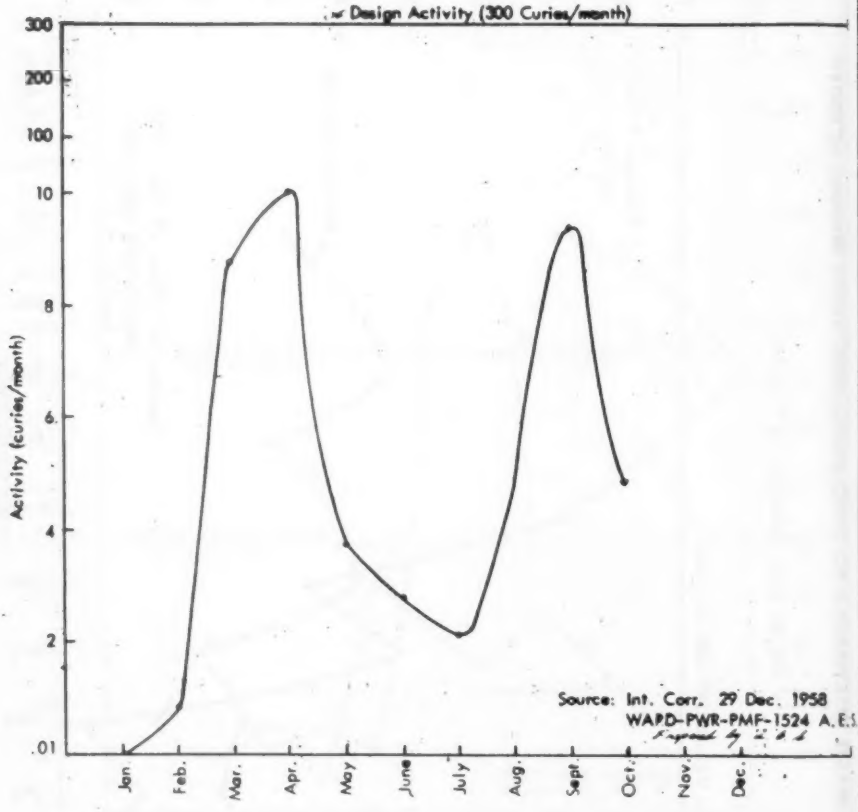


Fig. 4 SPECIAL WASTE DISCHARGED TO OHIO RIVER – 1958, GROSS GAMMA ACTIVITY

Fig. 5 TRITIUM ACTIVITY IN REACTOR EFFLUENT DISCHARGED TO OHIO RIVER - 1958



APPENDIX D

TRAINING ACTIVITIES
DIVISION OF RADIOLOGICAL HEALTH

The Public Health Service, within its Division of Radiological Health, conducts a training program at the Robert A. Taft Sanitary Engineering Center. The purpose of this program, instituted in 1951, is to narrow the gap between the acquisition of new knowledge and its application in the laboratory as well as in the field. This is accomplished through presentation of intensive short-term technical courses, specifically designed for professional personnel from regulatory agencies, industry and academic institutions.

Currently nine courses are offered. Those related to radioactive waste disposal include:

a. A two-week course in Radiation Protection dealing with in-plant aspects involving the handling of radioactive materials. Hazard evaluation methods are introduced and proper handling and disposal procedures are stressed.

b. A ten-day course, Environmental Health Aspects of Nuclear Reactor Operations, stressing the importance of site selection and waste management in minimizing potential health hazards.

c. A one-week course, Radioactive Pollutants in Water, directed toward water pollution control personnel and covering waste disposal and water surveillance methods.

d. A one-week course, Radioactive Pollutants in Air, similar in scope to the water course, but directed toward air pollution control personnel.

e. A two-week course, Determination of Radionuclides, related to the radiochemical analysis of waste materials and environmental media.

f. A two-week course covering the sanitary engineering aspects of nuclear technology, designed to meet the need by key personnel in administrative and directive capacities for broad knowledge in nuclear science and technology.

These courses are conducted by a permanent professional staff augmented by personnel from Public Health Service headquarters and research programs and consultants from universities and other federal government agencies such as the AEC, USOS, and NBS.

During the past 3 years, nearly 500 persons have attended the courses described above. Over 10% of these trainees have been from industry.

To these might be added an almost equal number of trainees attending similar courses at the Robert A. Taft Sanitary Engineering Center in the fields of air pollution, water supply and water pollution control, and milk and food sanitation. These latter courses are sponsored by the Division of Sanitary Engineering Services. Although a detailed study of radioactive waste disposal is not their principal objective, the problem as related to each specific field is incorporated into all such courses.

Many laboratories at the federal, state, and local level are today analyzing environmental and waste samples for radioactivity. Recognizing the need for evaluation of data obtained as well as techniques employed, the the Division of Radiological Health supports, in cooperation with the Division

of Sanitary Engineering Services, an Analytical Reference Service. Through distribution of samples, containing known concentrations of radioactive materials, to some 50 member laboratories, the Analytical Reference Service has obtained valuable information in this realm. Such information is of considerable significance in evaluation of current waste disposal methods.

Mr. TERRILL. In view of the comprehensive nature of these hearings and the excellent presentations made by the other speakers, my remarks will be confined to those aspects of radioactive waste disposal that seem to be of major importance from a public-health standpoint. In addition, I will comment on economics of waste disposal principally as it may affect health agencies and purveyors of public water supply.

From the public-health standpoint, the primary consideration is the overall effect on people of exposure to all types of radiation in the expected but variable pattern of sickness, health, and age. Consequently, we must bear in mind that radioactive wastes are only one facet of possible radiation exposure that must be considered by health agencies. Therefore, in determining criteria for the safe disposal of radioactive wastes, all means of exposure of a local or national population must be considered.

In view of these principles, it is most important to know the current concentration of radioactive materials in air, foods, drinking water, and the existing protective mechanisms. Soon after the creation of the Atomic Energy Commission, the Public Health Service developed a research program cooperatively with the Commission, the Department of Defense, and with the advice of several professional organizations, to learn quantitatively about radioactive wastes, and the effectiveness of existing water-treatment facilities.

The results of the early stream studies have been presented by Dr. Tsivoglou in his reference to the Columbia River and the Savannah River studies. Concurrently experimental work on decontamination of radioactive waters was undertaken at Oak Ridge. The results of these studies may be summarized in the following way:

In the water-treatment processes involving coagulation, settling, and filtration, most of the radioactive particles will be removed, but only 10 to 70 percent of the dissolved radionuclides will be removed. Ion exchange and distillations which are more effective are also much more expensive. Thus first consideration should be given to the control of radioactive wastes at their source and second to alternate water supplies, preferably from ground-water sources. Environmental surveillance activities indicated that the waterworks operator and the health official were not only confronted with natural radioactivity and possible waste materials from reactors and radioisotopes, but that very appreciable quantities of gross radioactivity from atomic weapons tests were contaminating water sources.

This background material defines at least qualitatively the impact of radioactive wastes, and associated activity, on both the industry and the public-health agencies responsible for the safety of water supplies. Thus, it now appears that the atomic-waste treatment facilities should not be allowed to discharge wastes which reach this Nation's waters unless they produce an effluent of sufficient purity so the water at the intakes of water-treatment plants will be safe from a radiation viewpoint taking into consideration natural and man-made background.

The waterworks and the health agencies should initiate evaluation to determine if raw waters are contaminated. If relatively simple techniques fail to assure a safe water supply, those responsible are confronted with major changes in their basic water-supply planning, either from the standpoint of sources or treatment or both. Estimates

of the cost of such major changes in water-supply systems, even on a single river system, would be a major engineering effort.

Thus the economics of waste disposal involves both the industrial unit producing the waste and the agencies responsible for the safety of public-water supplies.

A few years ago, the resources of the Public Health Service were shifted to activities which would have the most immediate impact on the evaluation of radiation emanating from power reactors. These include planning with the designers and the measurement and evaluation of all the radioactivity which is found in the environment. An opportunity for both study, service, and experience became available during the planning of the Shippingport reactor. It was our belief that this reactor system could serve as a prototype for the development of a satisfactory technical and administrative operating evaluation system of a typical power reactor. Through the cooperation of the Naval Reactor Branch of AEC, a quantitative report on prospective waste, an evaluation of the waste treatment system, and the probable effects of the treated waste on the river were developed.

With the State of Pennsylvania and the Pittsburgh Operations Office, an administrative instrument in the form of a waste disposal permit was developed which was approved by the State board of health. This document serves as the basis of both administrative action and public understanding. The Public Health Service has continued its assignment of personnel to the Pittsburgh Operations Office, and that agency has continued to supply data which illustrate the type of information which public health agencies must tabulate and analyze to assure against overexposure.

The prospective operator of nuclear reactors is confronted with a complex environmental situation in which the radioactivity being discharged to the environment is mixed in with other natural and man-made activity already present in similar concentrations. While these concentrations are low, they are in a range that is around a level of activity of 10 to the minus 8 microcuries per ml (10^{-8} μc/ml.) which has been considered to be the level at which further identification of radionuclides is necessary to assure public exposure below maximum permissible levels.

At this point, I should like to depart from my prepared remarks to say that I feel it is very important for the designers and operators of nuclear powerplants to work closely with local, State and interstate health agencies in matters involving radioactivity. In this regard, Admiral Rickover and his staff have been exemplary in cooperating closely with us both at the Shippingport atomic power station, and in connection with the operation of nuclear-powered naval ships.

They have provided detailed technical information which clearly indicates a great deal of care in the design of their waste disposal operations and provides a sound basis for mutually checking conditions. The same thing has been true, I might add, of the Maritime Commission in their program.

Representative HOLIFIELD. Do you find in your work around the Shippingport plant that you have established liaison between the Public Health Service and the Shippingport people and the AEC, and is there a coordinated program there in which you take into con-

sideration all types of environment, other than that which is produced by the Shippingport plant?

Mr. TERRILL. Within the bounds of our current knowledge and the more complex biological and ecological systems that were referred to by Dr. Morgan, yes.

Representative HOLIFIELD. There is an area that has to be coordinated; is there not?

Mr. TERRILL. Yes, sir.

Representative HOLIFIELD. This depends a great deal on the evolution of a body of law which will assign the responsibility more definitively to the people that are responsible; does it not?

Mr. TERRILL. It depends upon that to a great extent, but it also depends on the existing protective systems we have further down the river in the form of water treatment plants in case an accident should happen. It depends on both.

Representative HOLIFIELD. The Public Health Service is in charge of the cleaning of the water down the river?

Mr. TERRILL. Yes, sir; on an interstate basis; the State is responsible for the intrastate portion of the stream.

Representative HOLIFIELD. What kind of burden is going to be put upon the Public Health Service in the way of additional responsibility because of these reactors that are being built? Is it going to be a substantial burden?

Mr. TERRILL. It would appear that the exact amount is a function of the rapidity with which State and local agencies can build up their technical competence. It would be a considerable burden in any case.

Representative HOLIFIELD. This will undoubtedly in time require additional appropriations for the Public Health Service and an additional complement of skilled people; will it not?

Mr. TERRILL. It would, but we would also hope that we could, through training, keep this requirement for appropriations, transferred to some extent to the State agencies, and over the long period that they would carry most of the burden.

Representative HOLIFIELD. In the event that the States do not come forward with their share of the supervision, appropriations, and training of competent people, will the Public Health Service assume this responsibility in the interim period? In other words, do you have an overall protection of the public from all types of radiation to the extent that you will fill the gap until State bodies assume this responsibility?

Mr. TERRILL. We will do our best to fill that gap; yes, sir.

Representative HOLIFIELD. What responsibility do you have to bring to Congress' attention the areas which are not properly covered by State, county and city governments?

Mr. TERRILL. We hope to do this through your committee and also through our appropriations hearings and through the congressional committees that we normally work with.

Representative HOLIFIELD. The field of industrial supervision and scrutiny is much narrower than the overall field of radiation hazard?

Mr. TERRILL. Yes, sir.

Representative HOLIFIELD. The Public Health Service at this time is the body which has been given the overall responsibility, although specific statutes define certain areas of Federal responsibility.

Mr. TERRILL. That is pretty much the case; yes, sir.

Representative HOLIFIELD. The Public Health Service being Federal, and the AEC being Federal, is there any conflict between the two in the discharge of this responsibility? Maybe I should put it this way: Is there any planning being done in a conference between the top people in Public Health and the top people in AEC as to solving this problem?

Mr. TERRILL. Yes, sir. Meetings have been held, and I understand that the Bureau of the Budget is undertaking steps to look into this in an overall way.

Representative HOLIFIELD. Have there been interim committees established by Dr. Burney and Chairman McCone to bring this matter together into a discussion and solution?

Mr. TERRILL. Yes, sir. There have only been a few formal meetings, but there have been a number of meetings at the technical level. There have been exchanges of correspondence and draft documents.

Representative HOLIFIELD. You may proceed.

Mr. TERRILL. With regard to the high-level wastes, I will only touch on this lightly because this has been covered so well by the previous speakers. However, I would like to mention that health agencies are vitally interested in the high-level waste research, particularly regarding the related health hazards involved in each system of transportation, chemical processing, and ultimate disposal. Operationally the health agencies understand that tank storage is the present method of choice for high-level waste. These storage facilities are now located at a relatively few Government-operated processing plants. In our planning it is expected that this situation will continue and that the economic burden of this phase of the waste disposal problem will largely rest on the Federal Government during the immediate future.

Limited staff work indicates that the cost estimate of high-level waste disposal in future years reported by Culler and others are quite reasonable, and that the Nation can expect that the operational requirements, related to the handling, transportation, and disposal of high-level radioactive wastes will require multi-million-dollar expenditures. The portion of this expenditure that will be placed on health agencies, local communities, and individual industries will depend on many factors, including acceptable health standards and the extent to which major changes in nuclear fuel and reactor operations develop, as well as factors related directly to waste disposal.

Representative HOLIFIELD. At this point, Mr. Terrill, will you pick up your summary, and we have already accepted your full statement. We are running short of time.

Mr. TERRILL. In summary, I would like to say that the principal matters of interest to us are related to methodology and health standards so that we actually know as well as we can what radiations are reaching people, what these effects on people will be, and that the nuclear energy industry will not be unnecessarily restricted either by calculations or standards that are based upon extrapolated data.

Representative HOLIFIELD. Thank you very much. Are there questions of Mr. Terrill by members of the committee?

Mr. HOSMER. I have one question. In your printed summary on page 13 you make the statement, "It is our current belief that the

overall radiation in the environment in the country will become more complex and the radiation levels will generally tend to increase to the point where current health standards will be threatened."

If that is taken out of context with the rest of your statement, and that of the previous witnesses, it would need a little explaining.

Mr. TERRILL. I think, for purposes here, I would merely like to remark that these current health standards are being changed, and changes are under consideration in line with what Dr. Morgan had to say. We don't know then precisely what the medical exposure is. Indeed in many cases we don't know what the natural background is. It is a continuing question as to whether or not all of the complex factors, including continuing or not continuing the foreign and other weapons tests will lead to apparent excessive radiation exposures. At the moment, I am not thinking of standards in a biological sense. I am thinking of standards with reference to calculations and the mathematical factors that people may add, such as a tenth or a hundredth to the standards primarily developed for occupational reasons. It is quite easy to get readings in this way in excess of calculated standards in some instances.

Representative HOSMER. In short, the quotation I read is a possibility we should be watching for rather than in the nature of a prediction.

Mr. TERRILL. Yes, sir. I would hesitate to predict this. I would also hesitate to predict that we won't see numbers that would be exceeded upon examination in an environment.

Representative HOLIFIELD. Thank you, Mr. Terrill.

Mr. TERRILL. Thank you, sir.

Representative HOLIFIELD. Our next witness is Dr. Abel Wolman, of Johns Hopkins University.

STATEMENT OF ABEL WOLMAN,[5] JOHNS HOPKINS UNIVERSITY, BALTIMORE, MD.

Dr. WOLMAN. Mr. Chairman, I was not too clear in my mind as to what the nature of this general summary was to be. I took the liberty of trying to look at the hearings since their beginning, with the idea of determining at least for myself, and I hope for the value of the committee, as to what the hearings up to the present moment seemed to disclose, at least in the highlights of the problem. Unless you have some serious objection to that, I should like to pursue that intent, and be interrupted at any stage if I seem to be going beyond that intent.

Representative HOLIFIELD. Go ahead.

Dr. WOLMAN. I was interested in determining as well as I could and on as simple a base as I could what we were talking about over the last 3 or 4 days. For the purposes of my comments, I should like to go back to stress that we are in fact discussing, to a degree never before presented, the types and the amounts of waste encountered in this industry, and likely to be encountered in the immediate and remote future.

They fall into two classes, which if not sharply delineated, at least do have major differences in their nature, in their volume, in their hazard, and in their control.

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[5] See biographical sketch, p. 7.

The first of these is the low-activity materials which have been described, and for which solutions at least at the moment have been considered to be satisfactory.

We have discharged at an annual rate approximately 9 billion gallons of low activity materials and with a total aggregate up to the present moment of 2½ million curies. We have a second category of wastes, quite distinct from these, both volumewise and strengthwise, which have been described as the high-activity wastes, with curies running to one or hundreds per gallon or plus.

In 1959 some 65 million gallons of these high activity materials are in storage and by various estimators the figure may run as high in 1980 to some 10 billion curies. By the year 2000 some 200 million to 1 billion gallons may have been accumulated.

I make the distinction between the two and then I want to return to the problems created by virtue of the two orders of waste, or the two sources of waste. It has been pointed out that the significant differences in control in relation to the two groups are roughly as follows:

In the case of the low activity or intermediate level activity waste, the program has been, and it has been a successful one up to the present time, to reduce the volume of these wherever possible with economy, and then disperse to nature under a very detailed control and management. I would not like to dispose of the problem of the low-activity wastes quite as simply as that, because if the industry develops in size, and in turn in increased production of waste, it will be confronted with a continuing necessity of the management of low-activity waste which will not be as simple as perhaps it sounds on paper. Volumes will increase, total curies will increase, and somewhere along the line, by some public responsibility, by some degree of total supervision, nature will not be so abused by these increasing volumes, even of low-activity materials that we will regret the result.

My point with respect to low-activity waste is simply that it is not a disappearing problem. It is a mounting one. Its great value is that it can be managed with economy, but it must be managed with rather careful criteria and extremely careful public supervision.

May I return to the high-activity waste problem. There, as I pointed out in the initial statement, the practice today is to reduce it in volume, if possible, to contain or hold, and to proceed with as much research and development, about which you have heard, in order to reduce that even further in volume, and also to put it in such state that it can be handled and controlled for indefinite periods in the future. I do want to stop a moment to evaluate, if I may, where it seems to me the testimony now leaves us in the disposal of high-level or high-activity materials.

A number of procedures have been suggested here. My evaluation, if I may dignify it as such, is a strictly personal one. It may have some value to the committee in indicating what next steps there ought to be. Much emphasis is naturally being given to fixation of such high-activity materials, either by ceramic methods at high temperatures, by sintering, by the fluidizing bed, or by direct chemical fixation which has not been too well described during these hearings. All of these are in a stage varying from laboratory demonstration to almost pilot-scale operation.

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In the case of the fluidizing beds, I would say it is in the second category of pilot plant operation. All of them give some promise. None of the testimony has indicated at this stage what the economics of this kind of fixation program will be. That remains to be demonstrated.

We have a second series of managing materials that at the moment are being held in tanks, namely, deep well injection, and disposal in selected geological strata, with the dominant emphasis on the probable use of salt beds. These methods are still in the highly experimental, almost semilaboratory stage. Their economics have not been evaluated, and it would perhaps be some 3 to 5 years before they can be. I do want to comment on utilization once again, because of the excellent summary statement by Dr. Manowitz, which I think supports my initial comment that utilization does not offer an answer to the waste disposal problem of high-activity material. It seems unlikely that it ever will.

The degree of purification which may be required is of such tremendous high efficiency that we know of no procedure by which it can be attained, certainly at this time.

On disposal at sea, Dr. Revelle will have more to say. I had the benefit of reading Dr. Pritchard's comment, and I would say that the enthusiasm for disposal even in coastal waters is quite low. The enthusiasm for ocean disposal is even lower. Unless Dr. Revelle comes forward with an indication that he is in complete variance with this, this is my present assessment of that potential for high-level material.

Transportation problems, of course, loom up large. They indicate some kind of continuing direct governmental responsibility for criteria of transportation, and certainly for supervision.

All of this would lead me to say that we are confronted with a necessity for a long-term research and development program. By long term, I would suggest that this may extend into generations. I make that emphasis because a short-term budgetary aid to research and development in this very important field, which extends over a year or only 2 years, is not likely to produce the result required. This is literally a long-term necessity for which there ought to be a continuity of appropriation which can be used by the Commission, by research institutes, by the various operations offices, and by all groups who may interest themselves in this.

I might say, in summary, with respect to cost that the costs of present handling of wastes is high in absolute figures. It is, as has been pointed out, relatively low in figures in relation to total potential nuclear fission power costs. But they are still higher than anyone would like to see them. Holding in tanks for any indefinite future is not attractive to most people in the business. Most would consider it as an interim necessity while these other developments proceed, we hope, at high pace.

May I say a word as to the implications of all of this. This is a struggle betwen protection of a human society and economics, in which the accomplishment has been high in protection and in which the economics have been correspondingly high in absolute numbers. The implications are there will have to be provided as time goes on a very high sustained quality of public supervision for the rest of the life of the development of the industry. Some comment has

been made in almost all the papers on the location of that supervision. My own judgment would be that it will not be an either/or kind of governmental supervision. This would seem to be not a contest between the State and the Federal Government, nor a contest, incidentally, between the Atomic Energy Commission and the other Federal agencies. There has been some suggestion that the knowledge, the equipment, the understanding, the deep-seated technology are resident only in the Atomic Energy Commission. I am sure that the Commission would be the last to accept that "exclusion" responsibility. My point is that the control of this industry in the future is a private-public combination. It is furthermore on the Federal level a responsibility of multiple agencies. I would predict that responsibility resting solely in the Atomic Energy Commission for inspection of this advancing nuclear industry would fall of its own weight if the industry grows as we hope it will.

It would appear to me that therefore this problem pushes us toward a division of understanding and responsibility by multiple Federal agencies. Perhaps, much of such a responsibility, in the long history of public health responsibility, is in the Public Health Service. Almost nothing that has been done in the atomic energy industry with respect to many, many methods of disposal of low activity and high activity wastes has been done solely by the Atomic Energy Commission alone. It has sought and has had the cooperation of the U.S. Geological Survey, the Public Health Service, the Weather Bureau, and many other Federal agencies. It would be my guess that this multidiscipline approach, multi-Federal-State agencies approach, will increase rather than diminish.

I might add, Mr. Chairman, that by the widespread development of the industry, a joint cooperative effort by Federal agencies and State agencies, to me at least, seems inevitable. If we go beyond the immediate problem of the nuclear power industry, it is incredible that the inspectional service of every source of radiation could be handled from a Central Government unit alone.

This becomes, therefore, not a choice, as I say, of one governmental level versus another, or of public responsibility versus private responsibility. The future will disclose that this will move forward on multiple fronts rather than on a single front. I say this with perhaps a greater degree of assurance than I might, but it is predicated on the fact that as soon as this total radiation industry of any kind falls into the normal streams of activity of our country, to my mind the better. When it does, it will fall under the competence, the review, and the supervision and inspection of the normal group of agencies with which our country has been accustomed to live for well over 150 years.

Representative HOLIFIELD. Thank you very much, Dr. Wolman.

Dr. WOLMAN. Thank you, sir.

Dr. RODGER. Dr. Wolman, I think you have made an excellent and succinct summary of what has gone on so far. I think there are two points which we might perhaps like to bring out in this summary on which you might wish to comment. One is the fact that the low level waste must inevitably always exist, and while the level that we permit ourselves may go up or down, depending on the growth of the industry and the variance of permissible levels as we learn more and more,

there will also have to be this use of our environment as a diluting medium. That is extremely important to the industry, as a matter of fact. One of the things I think we should emphasize is that one of the very big reasons for hanging onto all of the high level waste is that a small amount of that uses up a large quantity of this diluting power, and we must hang onto that jealously because it is important to us in the operation.

Dr. WOLMAN. May I summarize my comment to this extent? I would say for well over 40 years I have been a strong advocate, in the case of other pollutants more familiar to us, of using nature, under control, as one of our major methods of disposal. I would establish and use exactly the same thesis with low level activity in the radiation field. I think we would be discarding an extremely valuable natural asset if we did not make use of nature with low activity waste. My only comment was that, when we do that, as we do in any other kind of pollutant, we must do it with logic, with criteria, and with persistent monitoring. That was not intended to indicate that we should stop doing it, because I think we would throw away an extremely valuable disposal method.

I share your view completely that we cannot do this at all with high level activity, because we would ruin the whole business.

Dr. RODGER. The second point is tied in with it somewhat, and has to do with costs. When we speak of the very high level of cost, we have a tendency, I think, to be thinking in terms of the high level waste and the cost of hanging onto it. I think we should emphasize the fact that the low level waste operates in general at a relatively modest cost. I think from a research institution such as our own we spend less than 1 percent of our operating budget in handling low level waste. I think this is true of the production sites, too.

The point here is that while this operates chiefly on the large volume, if we so contaminated our diluting environment that we had to treat the entire waste, then the statements which are sometimes erroneously made, that waste disposal is going to put us out of the economic picture, would be true.

Dr. WOLMAN. I am sure it would be true if we had to do that with low level activity. I keep referring to the high level activity waste cost as high in unit absolute values, and low in relative values. I would stress that fact. I would add this comment. If we were to rehearse in any of the familiar industries the cost of waste handling, I believe we would get ourselves into a similar pessimistic situation of saying that absolute cost is pretty high, relative cost in the unit product, either of steel or paper or anything else, is quite low. I think it is worth stressing that. We move in this field from an optimistic level to a pessimistic level all the time. Optimism is generally characteristic of the operating man because he is close to the task and he sees it as a family type of activity, sometimes perhaps with a higher degree of optimism than I would share. The pessimistic regulator or supervisor looks at it from the other end. Somewhere in between is probably the route we should travel.

For example, when Mr. Sporn points out that the handling of high activity wastes will ultimately have to become an industry responsibility, a feeling which I share—ultimately, and he compares it with the wastes with which they have struggled over the years, namely, the use

of fossil fuels and the handling of coal ash, and the like, I would merely point out that he looks at it from the optimistic standpoint, while the industry, with the possible exception of his own group, has not been eminently successful in keeping a good deal of that ash out of a good deal of the atmosphere or certain other areas.

Representative HOLIFIELD. This would apply to the pollutions in the streams with chemicals, too.

Dr. WOLMAN. Without question. I would reduce the total optimism of some of the hearings, and I would raise the total pessimism of some of the hearings to what I think would be a more realistic picture of the world in which we live.

Representative HOLIFIELD. On the point of the State compacts, the upper and lower States of the upper Colorado have been engaged in litigation for the last 29 years as a result of compacts, and have not arrived at a solution yet of the division of the water of the Colorado.

Dr. WOLMAN. I should not whisper this, but in all of the activities the progress generally is geological and not chronological.

Representative HOLIFIELD. Thank you very much. At this time we will ask Dr. Roger Revelle, of the Scripps Institution, to come to the stand. We are going to try to conclude the hearings even at the risk of running over a little bit, because of conflicting appointments of some of the members during the afternoon.

Dr. Revelle, we are glad to have you with us.

STATEMENT OF DR. ROGER REVELLE,[6] SCRIPPS INSTITUTION

Dr. REVELLE. I take it that you want me to talk particularly about the possible effects of the introduction of waste into the marine environment.

Representative HOLIFIELD. Yes.

Dr. REVELLE. I would first like to enter a caveat that our knowledge of most of the processes in the ocean is altogether too fragmentary to permit precise predictions of the results from introduction of a given quantity of radioactive materials at any particular place. In order to obtain the necessary knowledge, adequate, long-range programs of research on the physics, chemistry, and geology of the

[6] Scripps Institution of Oceanography. Date and place of birth: March 7, 1909, Seattle, Wash. Education: A.B., Pomona College in 1929; Ph. D. in oceanography, University of California in 1936. With the exception of service in the Navy during and immediately after World War II, his entire career has been spent at the University of California's Scripps Institution of Oceanography, of which he became director July 1, 1951. Now director of the University of California La Jolla campus and of the Institute of Technology and Engineering, University of California. On staff of Adm. W. H. P. Blandy in Operations Crossroads in charge of oceanographic measurements. Organized resurvey of Bikini in 1947. Fellow of American Association for Advancement of Science, San Diego Society of Natural History, Geological Society of America; member, American Meteorological Society, Sigma Xi, Society of Limnology and Oeanography, Geological Society of Washington, Western Society of Naturalists, Cosmos Club, Men's Faculty Club, Berkeley, American Association of Petroleum Geologists, and American Geophysical Union. President for 1956–59 of the section on oceanography of the American Geophysical Union. President of special committee on oceanographic research of International Council of Scientific Unions; chairman, National Academy of Sciences Committee on Biological Effects of Atomic Radiation in Oceanography and Fisheries; member, National Academy of Sciences—National Research Council Committee on Oceanography; member for North America of International Advisory Committee on Marine Sciences of UNESCO; member, Committee for the International Geophysical Year of the International Association of Physical Oceanography; the Panel on Oceanography of the National Committee for the International Geophysical Year; and Divisional Committee for Mathematical, Physical, and Engineering Sciences of National Science Foundation. In 1954, Dr. Revelle received the Albatross Medal of Swedish Royal Society of Science and Letters for outstanding achievements on oceanography, especially in deep-sea research. Honorary doctor of scienc from Pomona Collge. Elected to National Academy of Sciences April 23, 1957, and to American Academy of Arts and Sciences in 1958. (Submitted by witness.)

sea, on the biology of its contained organisms is required. In addition to engineering studies of particular waste-disposal problems as they arise. The results of engineering studies are necessarily limited in application. Moreover, they must always be conservative, at least until sufficient broad understanding is obtained. It is also well to keep in mind that all parts of the ocean and its bordering seas are in communication with each other, and are in continuous motion, although it is true that the rates of movement differ greatly in different areas. There is eventually complete interchange of water between all oceans and seas, some parts being partially isolated from others, and the exchange between these parts being much slower than within them.

What happens in any point in the sea ultimately affects the waters everywhere. Of course, the oceans are international. No man and no nation can claim the exclusive ownership of the resources of the sea. The problem of the disposal of radioactive wastes in the ocean, with its potential hazard to human use of marine resources, is an international one. As I pointed out at previous hearings, it is one of the subjects of concern to the International Atomic Energy Agency.

The comments that I will make are primarily a summary of the conclusions that have been reached by two National Academy of Sciences-National Research Council committees: the Committee on the Biological Effects of Radiation on Oceanography and Fisheries, and the Committee on Oceanography. I am a member of the Committee on Oceanography, and chairman of the Committee on the Effects of Radiation on Oceanography.

The attractive thing about waste disposal in the ocean is that the ocean represents a great hole in the ground. The volume of water is tremendous. Dispersal of at least low-level radioactive wastes can take place relatively rapidly down to a level at which these materials can hardly be detected. Moreover, by putting materials on the bottom in containers, which hopefully can remain intact for periods of the order of 10 years, we can also achieve the other consideration of waste disposal; that is containment, or isolation from human beings and their foods.

It is also true that this containment, that is, isolation, can probably be obtained by disposal of liquid wastes in the deep sea. At the present time everybody who is concerned with the practical or engineering aspects of radioactive waste disposal is interested in the ocean only for disposal of low-level waste. It is possible, however, that in the future, some 30 or 40 years from now, at least in some areas, and with some kinds of high-level wastes, people will want to dispose of them in the deep ocean.

What are the potential hazards from the disposal of radioactive wastes at sea? If we put them in certain areas, the transport of the water may bring these materials onto the beaches and into swimming areas, and the people utilizing this marine resource, the recreational resource, may be subjected to radiation from the water and from the materials of the beaches which tend to absorb radioactive materials.

A far more serious potential hazard is the contamination of marine foods. All marine organisms tend to concentrate radioactive materials just as they concentrate other elements and compounds from the environment. This concentration often is by factors of a thousand

or more over the concentration in the environment. It is for this reason that the maximum permissible concentration in any large volume of sea water has to be much lower than the maximum permissible concentration in drinking water. It is simply because the marine food organisms act as little chemical processing plants and absorb the materials.

For example, with phosphorus, the maximum permissible concentration in a large volume of sea water is probably one twenty-five-thousandths what it is in drinking water. With strontium 90, one-half to one-third the drinking water concentration should be permissible. With iron 59 and cobalt 60, probably a factor of 1,000 less than drinking water. With iodine 131, I might say, it is only a factor of 10 less than drinking water. With cesium, it is a factor of 20.

One other interest of man in the introduction of radioactive materials in the sea is the possibility of harmful effects on the marine food resources themselves, both somatic and genetic effects. There is very little known about this. It is generally believed to be a negligible importance; that is, the principal potential hazard is assumed to be the transmission of radioactive materials to people through marine foods. This potential hazard is not of very great consequence to most people in the United States, because our consumption of fish and marine food products is only about 10 pounds a year per person. But for other peoples, and for some people in our coastal regions, the potential contamination of marine foods is a more serious matter. These are the people who get virtually all their protein, a good deal of their calcium, and therefore a good deal of strontium from marine foods. Such people would tend to eat about 3 or 4 pounds of fish or marine invertebrates a week.

Mr. RAMEY. Would that apply to Japan?

Dr. REVELLE. Yes. For example, in Japan, virtually all the animal proteins that the Japanese obtain are from fish and marine invertebrates.

Mr. RAMEY. Of course, they get less fallout.

Dr. REVELLE. That is right. The fallout problem is much less severe from the Japanese point of view than our own because of the fact that the fish get so much less strontium in proportion to the amount of calcium than our milk cows do.

There are three areas that we should keep separately in mind in thinking about the disposal of radioactive materials in the marine environment: Estuaries, the inshore environment, and the offshore environment of the high seas. On the United States east coast, the estuaries are of the kind in which the bottom water moves inshore. So there is a tendency for radioactive or any waste materials to remain—the materials disposed of on the bottom—in the estuary and not be dispersed. These harbors and inshore waters in general also contain many bottom-living animals and many bottom-attached plants. These concentrate radioactive materials in some cases by factors of more than a thousand. So that the dispersal or removal of radioactive materials from these harbors and estuaries takes place very much more slowly than the dispersal of the water itself, or of other substances which are not concentrated by organisms, or which are not absorbed by the bottom sediments.

In coastal waters, that is waters within 200 miles of the coast, we can think of two different situations: the shallow coastal waters of the Atlantic coast of the United States, and the relatively deep waters which exist quite close to shore off the Pacific coast. The shallow waters also are typical of the gulf coast. Here there are possibilities for disposal of rather large amounts of low-level wastes.

Finally, we have the deep sea, which lies below a virtual screen or curtain called the thermocline, and is thereby isolated from the rest of the ocean. Very few of men's food organisms come directly or indirectly from the deep sea and it has the advantage of isolation from the part of the ocean that is of most concern to man.

What happens to radioactive materials put into the ocean? We can say basically three things. First, they are dispersed; that is, they are widely spread and diluted. They are brought down to very low levels of concentrations by currents and turbulence. Second, they are concentrated. They are brought to high levels of concentrations by marine plants and animals. Third, they are removed from the water and absorbed in the bottom sediments, and in the bottom-living organisms. This absorption into the bottom materials is, in almost all cases, a great safety factor for disposal in the marine environment. For example, at Windscale in the Irish Sea, the British are now disposing of an amount of radioactivity amounting to thousands of curies, perhaps as much as 10,000 curies a month. The reason that this is possible is partly because of dispersal of the material by the rapid currents of the Irish Sea, but apparently very largely because of the absorption of the radioactive materials in the sediments around the outfall.

The National Academy of Sciences committees that I have mentioned previously have appointed a working group which has just recently completed a report on the possibility of disposal of low level wastes in the Atlantic and gulf coasts. Professor Dayton Carritt of Johns Hopkins was chairman; the members of the committee are listed in my material for the record. This committee has recommended that there could be at least 15 disposal sites within 50 miles of the shore on the Atlantic coast and about 10 disposal sites within 100 miles of the shore on the gulf coast. They have recommended that initially no more than 250 curies per year, or 100 curies per month be disposed of in any one of these sites. The disposal sites should be not less than 75 miles apart. Before any disposal actually takes place, a survey should be made of the fishing activities in the region, of the sediments and their absorptive properties, and of the bottom living organisms—bottom living organisms which might be used for human food or which could serve as pathways by which the fish might take up radioactivity—and also organisms which could be used as chemical concentrators in subsequent studies of the area. Even when it is impossible to find radioactivity in the water, it may be rather easy to find it in animals living on the bottom, concentrated sometimes by a factor of several hundred thousand.

They have also recommended that any low-level disposal area on the Continental Shelf of the east coast should be monitored after operations start, to see what happens to the radioactive material, whether in fact it is dispersed to a level that is so low that it can no longer be found. (In the disposal areas used to date by the AEC this does seem to be the case. We are unable to find any radioactivity in the vicinity

of the disposal site.) It may be possible as a result of such monitoring and surveys of these disposal sites greatly to increase the amount of radioactivity that can be disposed of.

As I pointed out, the British experience indicates that literally thousands of curies, perhaps even tens of thousands of curies, can be disposed of rather close to shore. Our committee has recommended to begin with only about 250 curies per year of packaged low-level waste in each disposal site, but they felt they had to be very conservative until more information will be obtained from the surveys and from the monitoring.

Representative HOLIFIELD. Mr. Toll has a question.

Mr. TOLL. Those places that you have been checking on the coastline of the United States, how long have your investigations been going on?

Dr. REVELLE. These are not investigations made by us. You mean oceanographers in general?

Mr. TOLL. I may probably have worded it poorly. You said today it would appear that none of the AEC disposals has indicated any radioactivity in the area. Do you have any idea as to how long those disposals have been there and how many checks have been made?

Dr. REVELLE. The checks are certainly not adequate. The checks have been of a very preliminary kind. For one thing, it is quite difficult to think of proper techniques for doing this. In the second place, the navigational problem is not by any means solved. We are not sure that we are looking at just the right area. The area concerned may be rather small. In the third place, the surveys have not been continuous, nor have they been done on a systematic basis. I would say at the present time all we have are indications that the disposal in these coastal areas is probably perfectly safe. We cannot really say this with certainty until we have made more continuous and systematic surveys.

Representative HOLIFIELD. There are commercial companies that are going beyond the Continental Shelf and dumping these materials.

Mr. REVELLE. Yes; that is true. On the Atlantic Coast the Navy has also disposed of quite a bit of material.

Representative HOLIFIELD. Are these high-level wastes?

Dr. REVELLE. All low level.

Representative HOLIFIELD. Are they encased in concrete tanks?

Dr. REVELLE. They are encased in steel barrels filled with concrete.

Representative HOLIFIELD. What is the estimate of the life of these containers?

Dr. REVELLE. Most people say that a safe estimate would be 10 years.

Representative HOLIFIELD. By that time the half lives are down to a point where you feel there is no danger?

Dr. REVELLE. Except, of course, for such things as strontium, cesium, cobalt 60, and some of the other longer lived products.

Representative HOLIFIELD. Those would be included the low-level wastes?

Dr. REVELLE. Yes; they are included.

Representative HOLIFIELD. Small amounts?

Dr. REVELLE. Small amounts; yes. From the standpoint of the long-lived fission products and long-lived radioisotopes in general,

such containment does not really serve a very useful purpose. It certainly is very helpful for the short lived. In fact, it reduces the concentrations by a very large factor.

Representative HOLIFIELD. What is the depth of the ocean at the point where they are disposed of?

Dr. REVELLE. At the present time, most of the disposal has been taking place at depths of more than a thousand fathoms. The National Academy committee has recommended these disposal sites at much shallower depths, of the order of 25 to 100 fathoms. In other words, from 150 to 600 feet deep on the shelf.

I was just about to point out these are areas which are not fished, either because the bottom is rough or because they have been the site of disposal of explosives or other toxic materials in the past. The attempt has also been made in recommending these sites to avoid areas crossed by cables because the repairs or maintenance of the cables might involve breaking or bringing up one of the canisters. It should be remembered in thinking of this inshore disposal that the fishing effort is going into deeper and deeper water, and probably within a few years we can expect bottom trawling and dredging for commercial fisheries down to at least a thousand fathoms on the continental slope.

Coming to the possibility of deep ocean waters—4,500 meters or more—for the site of disposal of high-level wastes, I pointed out that the waters of the open ocean are stratified below the upper 100 or 200 meters, which is the area where all of the plant growth in the sea takes place and where most of the fish that we are interested in live. The zone of stratification is called the thermocline.

The surface waters are isolated, or partially isolated, from the deep waters by this boundary, this curtain of increasing density. Although there is no very clear theory on this, it is generally believed—and there is quite a bit of evidence for believing—that the deep water is isolated from the surface layer for periods of the order of hundreds of thousands of years—in the Atlantic for periods of a few hundred years and in the Pacific periods of perhaps 2,000 years. Moreover, there is reason to believe that the deep water is thoroughly mixed over this time period, so that any radioactive materials introduced into the deep sea in liquid form would be dispersed throughout the entire ocean. We have made some calculations on the basis of an estimate of the amount of fission products which would be produced per year by the year 2000, estimating about a thousand tons of fission products per year in that time. If this were put into the deep sea, it is not by any means impossible that this material would be spread out uniformly through the deep sea, and by the time it got up into the surface that the concentration of strontium 90, which is by far the most serious of all the fission products from the standpoint of this particular type of disposal, would be, roughly, 6.5 times 10 to the minus 8 microcuries per milliliter, or down by a factor of five or six below the maximum permissible concentration in inshore waters. It is probably down by a factor of 20 below the maximum permissible concentration for offshore waters. This is based on the calculations of uptake of strontium and calcium by marine fish and invertebrates and on the assumption that we are trying to protect the population which obtains all its protein and all its calcium from marine fish.

It should be pointed out that this calculation assumes an even distribution of the materials throughout the deep layer. It thus assumes rather rapid dispersal throughout the layer, which depends on currents and mixing, and it assumes a very considerable holdup time in the deep water. All of these are subjects which need to be investigated.

Representative HOLIFIELD. Dr. Rodger has a question.

Dr. RODGER. This matter of mixing or the lack thereof, isn't there some dispute among you oceanographers on that subject? In particular, didn't the Russians present a paper at the last Geneva Conference in which they disputed this point?

Dr. REVELLE. Yes. Their paper was really not pertinent to this point. A German oceanographer had suggested that some of the deep trenches (at depths of 8,000 to 9,000 meters or more) might be isolated from other parts of the deep sea, and the Russian paper pointed out that these trenches are in fact in free communication with other deep sea regions, which is what every reasonable man expected all along from the quite considerable measurements that have been made of the temperature gradients in the deep trenches. The waters of the trenches are not little pockets of stagnant water. They are in continual interchange with the rest of the deep sea.

Dr. RODGER. Then what you are talking about is not the very deep isolated areas, but the rather wide ocean floor at more moderate depths?

Dr. REVELLE. Yes, I am talking about the average depth of the ocean, which is about 4,000 meters. There are some isolated places, for example, right off Venezuela, called the Carioca Basin in which hydrogen sulphide is present in the bottom water. This is an isolated place. The deeper part of the Black Sea is also isolated for very long periods. So there are some quite deep water areas which are isolated.

Dr. RODGER. I saw a Polish paper on the Black Sea point which said that was not true.

Dr. REVELLE. A Polish paper?

Dr. RODGER. I believe it was a Polish paper.

Dr. REVELLE. I am sorry to say I have not seen this particular paper. There is quite a bit of evidence, however, that the waters in the Black Sea—the deep waters—may remain there for periods of the order of 4,000 or 5,000 years. This evidence is by no means conclusive, and I would like to look at this Polish paper.

Representative HOLIFIELD. How much additional material do you have?

Dr. REVELLE. I am all through this. I simply have some material to submit for the record.

Representative HOLIFIELD. You may submit anything you wish to supplement your statement for the record.

Dr. REVELLE. Thank you.

Representative HOLIFIELD. Because of the lateness of the hour, we are going to dispense with the round-table discussion at this time. However, tomorrow morning we are going to have a list of witnesses, and if we can get through with them, we will have a round-table discussion at that time. If we get time we will have a round-table discussion tomorrow, and any of those who wish to remain may partici-

pate in it. However, you are not requested to be obligated to remain. Mr. Ramey has a question before you leave, sir.

Mr. RAMEY. On your research program affecting ocean disposal, do you think it is adequately funded now?

Dr. REVELLE. No, I don't. I don't think so. This is prejudging the matter a little bit. The National Academy of Science Committee on oceanography will issue a report in about 15 days which will spell this out in more detail (p. 1324). Our general feeling is that for the overall problem of radioactive contamination of the oceans—this involves quite a few things besides radioactive waste disposals, such as explosion of nuclear devices, problems of contamination resulting from nuclear-powered ships, accidents to power plants near the beach, and all the other ways in which radioactivity can get into the oceans—our feeling is that in order to be able to protect the marine resources and at the same time to utilize the ocean, as Dr. Wolman pointed out, to the limit of its capacity to protect human beings from radioactivity, the research program should be rather considerably increased. In my material for the record, I have a detailed research program (p. 1299). If you will excuse me for a second, I will look up the exact figure.

We estimated that on an annual basis the total should be about $6 million. This represented about a $20 million increase over a 5-year period. In other words, about $4 million a year over a 5-year period above that which is being spent now. For this particular field, we feel that the marine scientists could profitably utilize about three times more than they are now spending.

Representative HOLIFIELD. Thank you very much, Dr. Revelle.

At this time we will invite Mr. Phillip Sporn, President of the American Electric Power Corp., to the witness stand, and he will give his testimony on the industrial viewpoint.

STATEMENT OF PHILLIP SPORN,[7] PRESIDENT, AMERICAN ELECTRIC POWER CORP.

Mr. SPORN. Mr. Chairman and members of the committee, I greatly appreciate the invitation to appear before this committee and address myself, as suggested by the committee, to the question of industrial participation and viewpoint. I shall confine my testimony to the point of view of the power industry.

For identification I am president of the American Electric Power Co. and American Electric Power Service Corp. In the filed copy of my statement for the record, I give a brief statement of my nuclear activity over the past 12 years, and I shall not repeat this information here except to point out that my current major activity in atomics is that of chairman of the Research and Development Committee of the East Central Nuclear Group, Inc., a group of electric utilities operat-

[7] President of American Electric Power Co. (formerly American Gas & Electric Co.) and of American Electric Power Service Corp. (formerly American Gas & Electric Service Corp.). Last 12 years or so has worked on various aspects of atomic power, particularly as they affect the power industry: as a present director and one-time president of Nuclear Power Group, Inc.; as a member for a period of some 4 years of the Visiting Committee for the Nuclear Engineering and Reactor Departments of the Brookhaven National Laboratory; as a member of the National Academy of Sciences National Research Council Committee on Disposal and Dispersal of Radioactive Wastes and as chairman of the Research and Development Committee of the East Central Nuclear Group, Inc. (ECNG), a group of electric utilities operating in the Ohio Valley and contiguous areas, who have joined together in a program of research and development which they hope will result in significant advances in the technology of producing commercially competitive atomic power.

ing in the Ohio Valley and contiguous areas, who have joined together in a program of research and development which they hope will result in a significant advance in the technology of producing commercially competitive atomic power.

The problem of atomic waste has obvious counterparts and problems arising in more conventional methods of generation where we must deal with the disposal of ash and effluents from stacks. The electric utility industry has been working on these latter problems for many years and has accumulated a valuable background of knowledge and experience. In the industry's fund of information and the Commission's knowledge and experience with atomic waste—if the industry's fund of information can be pooled in the public exploration of the problem—I am confident that practical solution can be found.

The hearings which are now being held under the auspices of this committee are therefore necessary and timely, and they should make an important contribution to hastening the peaceful and competitive use of the atom with safety.

Some prediction about the growth and sale of atomic power installations is necessary in order to get an idea of the size of the waste-disposal problem and of the timing of measures to cope with it. As a guide to my own thinking, I have been studying this matter intensively for some years. Without going into the details of these studies. I found it helpful in considering the developments in atomic power between now and, say, 1975, to break this period up into at least two intervals; one between now and 1965, and the other period between 1965 and 1975. The period after 1975 seems to me likely to present still different conditions.

If we examine the interval between now and 1965 from the standpoint of our industry, we find that the industry is going to be confronted with the need of installing then in that time some 100 million kilowatts of generating capacity. Since no reactor or reactor concept presently known gives firm promise of producing energy in the United States at a cost less than that produced by conventional fuels, it seems to me unlikely, speaking relatively, that very much of this generated capacity that will be built by 1965 will be atomic.

I have assumed that the prototype and full-scale atomic plants which will be installed in this period will be for the purpose of demonstrating the practicability of the several new and improved reactor concepts which are the subject of the country's research program. This might be of the order of 1 million to 2 million kilowatts.

The period between 1965 and 1975 in my view is going to be presenting a much more optimistic picture, atomic powerwise. In that interval it can be expected that the power systems of the United States will add around 200 million kilowatts of generative capacity. By 1975 the then total capacity will be of the order of 400 million kilowatts, and of that total, I believe that roughly 5 percent or 20 million kilowatts may be atomic.

More important, however, it seems to me, is the period beyond 1975. The chances are that in the 5 years following 1975, there will be as much as 110 to 125 million kilowatts of new electric capacity installed in the United States, and of this I think about 25 percent may be nuclear.

So in my own consideration of the problem I have in mind the volume of waste products that will be associated with the atomic-power installations in the several intervals I have studied. Within 20 to 25 years from now, under this view, we are in for an industrial development in atomic energy of as much as 30 million kilowatts in a period of 5 years, or at the average rate of 6 million kilowatts a year.

The rate in 1980 may well be 10 million kilowatts. If you assume that roughly one and a half kilograms of fission products will be produced for each electric megawatt generated with nuclear fuel, then by 1980, 75,000 kilograms of fission products will be produced a year.

These figures afford a basis for understanding the responsibility that is involved in this atomic-waste problem considered merely from the industrial angle. To put the problem into proper perspective, I should point out that under the assumptions I have made, the volume of atomic waste which will have to be disposed of represents a small fraction of 1 percent of the waste from fossil fuel plants which we have had to learn, and have learned, to handle. The quantities involved would offer no barrier if it appeared to be the best solution to store in relatively temporary leakproof vaults at plant sites.

Relatively temporary storage of this kind could go on for many years, but considering the character of the waste, such solution is hardly likely to prove good enough for the long term. It seems to me, therefore, we are in a difficult, long-term situation. We have a difficult problem to solve, and we have not very much to guide us in the way we are trying to make bricks without clay or straw. Yet there are some fortunate factors in this situation, and one is the following:

1. In dealing with reactors designed to generate power, we are dealing with something that is under rigid and tight control, and the system of safeguards that we have set up at the present time is certainly on the conservative side.

2. The problem is not going to become acute overnight. It seems to me that it takes something like two decades for atomic power really to get rolling in the power industry and therefore there is time—not time to squander—but I believe adequate time to work on these problems, and to bring them under effective control.

With this background, I would like to discuss briefly three questions which I believe are especially pertinent:

1. Where should the responsibility be for the disposal of waste products created by the power industry?

2. Will the development of reprocessing operations by industry enhance effective responsibility and the program of development of peaceful uses of atomic power?

3. In the field of government regulation, where should the ultimate responsibility—local, State, or Federal Government—lie?

Let me preface these questions by saying that I recognize there may be more than one position to be considered on each of them to the extent that different views exist. I understand that these hearings have been planned to bring out all of them. I state my own views without attempting to elaborate others which have been considered and rejected by me and which I have assumed other witnesses will cover.

The first question, where should responsibility lie for disposal of waste products created by the power industry? It seems to me that

if the atomic-power industry is to develop to as significant a point as I have indicated in my preliminary observations, it is most important that industry have as its responsibility the complete operation, starting with the planning of the reactor, working out all arrangements for it, licenses and contracting for the fuel, including processing and fabrication of the fuel elements, and the design and construction of the reactor from the very beginning, when the reactor first becomes critical to the very end of its life. Unless such arrangements are feasible and it becomes possible to work them out readily, it would appear to me that there is a great danger that the industry would be hesitant to go into atomic power on a major scale. If these conditions are not met, I fear in many critical cases industry may make antiatomic decisions just because the problems, the responsibilities, the costs and the economics of the operation could be clearly visualized in the conventional form and might not be clearly possible of visualization in the atomic form. It would appear to me that the atomic industry simply would not develop, or develop with the necessary virility, if somewhere in the total cycle a major governmental operation other than regulatory, even of the most benevolent kind, is introduced.

As a matter of fact, it strikes me that it is hardly in the public interest to let the industry shake the responsibility as comprehensive in the atomic field as it must assume in the conventional field. If industry is to be the medium for making available to the people of the United States the benefits of the atom in its power supply system, industry had better undertake to the fullest degree possible the complete responsibility.

In planning for use of atomic power for the normal long term needs of our power industry, it seems to me that we must bring it just as closely as we possibly can within the framework that we have employed so successfully in developing conventional power systems. I repeat that we are fortunate that we have time on our side in working out an effective program in which industry can undertake the complete responsibility I advocate. We should recognize the principle now so that our studies and our pertinent research and development programs will be conducted with this goal in view.

The second question, the development of reprocessing operations by industry, and hence effective responsibility in the program of development of the peaceful uses of atomic energy.

Senator ANDERSON. Don't you want to stop with the first one, Mr. Sporn?

Mr. SPORN. I would be delighted, Senator.

Senator ANDERSON. I am trying to figure what it means. Are you speaking this morning as an operator or did you speak as president of some east central nuclear group?

Mr. SPORN. No, I am speaking, Senator, strictly as a member of the power industry and as an executive in the power industry.

Senator ANDERSON. You say that if the atomic power industry is to develop—I am reading on page 8—to as significant a point as you have indicated in your preliminary observations—how significant was that point? You listed about 1 million or 2 million. As I count up what we now have in sight for 1961, it comes to about that. I would conclude you don't think there will be much done in the years

that follow 1961 or the present things that are underway, such as Consolidated and the Dresden and Shippingport.

Mr. SPORN. I have indicated 2 million possibly by 1965 in my testimony.

Senator ANDERSON. You said 1 to 2 million.

Mr. SPORN. Yes, sir. I think this is about what it is going to shake down to. As you know, Senator, some of the things we are talking about now won't go into operation for 2 or 3 years.

Senator ANDERSON. Yes. I want to see how much significance you did attach to the development. You say if those things could happen, it is most important that industry have under its responsibility the complete operation, starting with the planning of the reactor, working out all arrangements for it, licensing and contractual, for the fuel, including processing and fabrication of fuel elements and design and construction of the reactor from the very beginning when the reactor first becomes critical to the very end of its life.

When you say starting with the planning of the reactor, I assume that the Government is to have no part of that?

Mr. SPORN. Senator, I am not talking about the research or developmental phase at all.

Senator ANDERSON. That is part of planning a reactor. You can't segregate research and development from planning, can you?

Mr. SPORN. We cannot. In my testimony I indicated that although by 1965 a relatively small number of kilowatts are likely to come on the systems of the power companies, public and private, of the order of 1 million or 2 million kilowatts, my best judgment was that by 1975 the capacity would be up to about 20 million kilowatts. That is one-sixth of the total electric capacity we now have in the United States. That begins to be a very significant item, Senator, in my judgment. It is about that really I am talking. I am not talking about what we are going to do in the research and development stages.

Senator ANDERSON. That is post-1975. I wonder if you were not talking about the period before 1975. I thought that is what you were talking about.

Mr. SPORN. I was talking of a first phase up to 1965. Then I indicated there is also a third phase after 1975.

Senator ANDERSON. Yes, but you were dividing the two periods, present to 1965, from 1965 to 1975, and in that time, including these two periods, you believe that the planning of the reactor, and so forth, is solely the function of industry. Can you relate that to the Philadelphia Electric proposal where $23 million and $40 million will come out of the Federal Treasury? If I went to you tomorrow and said, "Let us build a power plant in some little community that you were very familiar with," you having great experience in the field, and I said, "You put in two-thirds of the capital and I will put in one-third, and I will plan it and run the show." Knowing you, I think you would say "No" pretty quickly.

Mr. SPORN. Senator, I think there is no inconsistency, and I hope by this time my views would be known to you in particular, because I have had the privilege of discussing these problems with you.

Senator ANDERSON. I thought they were until I heard this.

Mr. SPORN. Yes, sir. I didn't mean them to be inconsistent. In my mind, they are not. I am sorry, apparently I have not set myself

clear on the record. I am talking about atomic power when it begins to be a question of day-to-day planning and constructing atomic powerplants because the people of the United States will then need that power. The power systems of the country, public or private, will recognize their respective responsibilities and begin to plan that power, and in due course want to make decisions to build powerplants. There is going to be in my judgment, Senator, a long period of time when critical decisions will have to be made week in and week out, and month in and month out, by many people in the power industry. The decisions will be this next unit or series of units, involving on a particular system 200, 300, or 500 thousand kilowatts. Shall it be atomic or shall it be conventional fuel, either hydro, which is also a fuel in a sense, or fossil fuel, gas, oil, coal, because the conventional technology, Senator, is so highly reliable and informed, so to speak, that any responsible power group can make a rather careful evaluation and have solid ideas of what capital costs and operating costs are likely to be in the present and the prospective future. It is my judgment that it is important if atomic power is to progress that they have similar relatively solid information about what atomic power is going to involve them in. I am not discussing the situation over a period of years when either prototype or research and developmental installations put in entirely by power systems or more likely, which I think is going to be the case, by power systems both public and private, with an indispensible item of Government help. I am not talking about that phase at all, sir.

Senator ANDERSON. I merely wanted to get it clear for the record, because someone picking this up might misunderstand this. You say this problem is not going to become acute overnight. It seems to me it will take something like two decades for atomic power to get really rolling in the power industry. I would not quarrel with that at all. That is 1979. What you are talking about here in the way of freedom is the period after 1980. I think I find myself in complete agreement with you. But if you are talking about what happens between now and 1965, or 1965 and 1975, there are some other problems. I merely wanted you to say whether this related to the present situation, or is it the period we are trying to get to when it will be done by the power company itself.

Mr. SPORN. I am definitely talking about the time, Senator, that everybody who is working in atomic power is looking forward to, when atomic power will take its place as a solid item in the economy. It is no such item at the present time. It is my belief and conviction from the 40 years I have spent in the power field that the sooner we develop our program so that we continue to throw that responsibility on the power system—I am sure the Senator knows I am not talking about a particular segment of the power industry—the better off the country is going to be, and atomic power also, Senator.

Senator ANDERSON. I know, Mr. Sporn, that you have put those 40 years into good purpose and you have a good understanding of it. I did not want as a result of a position built up for yourself in the power industry that somebody would come out the next morning and say that Phillip Sporn advocates that the whole thing be turned over tomorrow to private industry.

Mr. SPORN. I have no such thing in mind. I would feel mortified if I were ever presented that way.

Senator ANDERSON. Thank you very much.

Representative HOSMER. Specifically on this waste disposal thing, however, we were told the other day it is of a continuing nature. Perhaps it is a custodial responsibility for the products long after the normal life even of corporations which have no limit on their life. What is your feeling on that particular point?

Mr. SPORN. I think I will bring out just a little later if I have a chance, Congressman, that there is room and need not only for government, but obviously for the Federal Government, also. As to that custodian nature job, I am not quite sure we can quite see it now. But I should not be surprised if that is the kind of thing that, if we need it, would develop. It is extremely fortunate that while we are pushing ahead with the development of atomic power that at industry and government levels, that we do have time to work out these difficult waste problems properly, to be sure that both the economics and the safety of the country are taken care of. In some respects not entirely visualizable at the present time. I think, therefore, it is entirely in order to make sure that we know in which direction we are trying to make the development. It is my conviction, and this is what I tried to say in that first question, that we want to work toward the end that the responsibility for that lies in the power industry, with all sorts of regulatory and licensing supervision that will be necessary, just as it is now necessary, and being taken care of in the conventional technology.

We have a great deal of clearance to go through before we can put into operation a high-pressure, high-temperature powerplant in almost all of our States.

Representative HOSMER. The only thing I am worried about is that here we have a somewhat semiperpetual custodial responsibility. In conventional development you don't have that. We are dealing with these very long-lived radioactive wastes, the radioactive life of which would in the normal course exceed that of any private enterprise.

Mr. SPORN. Yes. Just to take one example, Congressman Hosmer, if our technology should indicate that we have a solid basis of disposing of them through some ceramic method, and there is good reason for that, and we can develop ceramic forms that can be safely put into the ocean on a half-life basis that nobody would be hurt, not even ocean life, then I would think automatically by that single step of technological development, which is certainly being worked on, we would have taken off some of the major item of that continuing custodianship. We will have taken care of it in that particular form. I am not sure that it will be the final form of disposing of it.

The second question that I asked, with the development of reprocessing operations by industry, and hence effective responsibility in the program of development of the peaceful use of atomic energy, as I have indicated before, one of the things that has been too long delayed in the development of the peaceful use of the atom is the introduction of the economic factor. The economic factor is an extremely difficult one to introduce at the governmental operation. If the economic development of the country is to be enhanced by the use of atomic power, it is important that atomic power be economical. I would think that the omission of private enterprise or private industry

responsibility from so important a phase and cost as the disposal of the waste product would delay the introduction of the necessary economic factor. That could not help but have a retarding effect.

From that point it is essential to turn the job over to private industry.

Representative HOLIFIELD. What you are really saying there is that you are advocating a strong research and development program right now so that this will be feasible.

Mr. SPORN. Yes, sir.

Representative HOLIFIELD. You recognize it is not feasible at the present time?

Mr. SPORN. I do recognize it. I am familiar with some of the research and development through some of the work I did for a number of years at Brookhaven. I am all in favor of that, and any further extension of it that is necessary. As I have said, I think we have time. But we have not any time to waste.

Representative HOLIFIELD. Do you think that the appropriations have been adequate in this field, or should they be increased slightly?

Mr. SPORN. I have been out of contact with that particular source of activity for about 2 years. At the time I was on the visiting committee, Mr. Chairman, I thought we were going along at Brookhaven at a good pace. I would hope and believe that is still the case today. I do not know, sir.

One of the things to which we are going to have to devote a lot of study is precisely who should do the reprocessing. In most cases I should think it would be best done by separate fuel processing industry. In some cases it may best be done on site by the power producer.

What I have said in relation to the industry taking over the responsibility for reprocessing waste disposal is intended as the ultimate objective. I recognize that industry could not now take over this field on a commercial basis, and that the ultimate objective will have to be achieved on a gradual basis over a period of time. For the time being the Government must continue. I would hope that would be the answer, and indeed expand its efforts in the field of reprocessing waste disposal. These efforts of the Government should be carried out in such a way that industry is brought into the picture to as great an extent as possible, and such effort should be terminated when the technology and economics have developed to the point where industry is able to take over the full responsibility on a commercial basis.

Representative HOLIFIELD. Mr. Ramey has a question.

Mr. RAMEY. In the omnibus bill the committee recommended last year, which was enacted into law, they provided a greater long-term contracting authority to the Commission to contract with private organizations in the field of reprocessing whereby the Government could reserve time or quantity or some way, and thereby help make a private project feasible.

Mr. SPORN. I think this is a good idea. I am all in favor of it.

Senator ANDERSON. I just want to say there, I could not agree more heartily than what you have said about the problem of waste disposal.

Mr. SPORN. Thank you. In the field of government regulation, where should the ultimate responsibility lie, in local, State, or Fed-

eral Government? Where should ultimate responsibility lie in the broad area of government jurisdiction, that is, where government has to be introduced into the picture. I believe here that for most purposes the responsibility will best be discharged if it is placed in the State or local hands. I say this with some conviction because our experience in more conventional technologies but in their most advanced form, in the field of very high pressure boilers, has shown that the present situation in the United States where the code, inspection, and licensing approvals are handled on a State basis, with State boiler inspection laws and rules, gives, on the whole, the most satisfactory arrangement.

It is true that in that case there are a number of national codes or guides to follow. We have, for example, in boiler practice the ASME boiler code. But the use of that code for safety control is different in different States. While that might appear to be a disadvantage for any organizations that operate in a number of States, as my own company does—we operate power facilities in seven States—the reverse is actually the case. For example, in connection with developing extra high pressure practice in boilers over the period of the last 50 years, our experience bears out our conviction that it is advantageous from every point of view to have local or State control.

In one case, involving proposed valving changes on extra high pressure boilers, involving more or less similar institutions in three different States, it was possible to obtain a very early and quick decision in one of our States to permit proceeding with the pioneer step even at a time when the national code was at least ambiguous in this matter. Subsequently we obtained a similar decision in a second State. In a third State, one of the most highly industrialized in the country, we had more extended discussions and negotiations and eventually we obtained the board's approval also.

All this work with the several State agencies was desirable from our point of view. The initial pioneering step in the first State did not detract from safety, and it helped the economics. The especially close scrutiny that the third State board gave to the matter had the disadvantage of delaying introduction of practice somewhat in that State, but, on the other hand, had the advantage of giving us further assurance of solid judgment on the question of safety.

Senator ANDERSON. Mr. Sporn, could anybody reading that interpret that as a belief that State legislation is easier to get by than Federal regulation?

Mr. SPORN. This, again, would not be my view, Senator, because I believe this. The advantage of State regulation is that there are 49 States. You therefore have an opportunity to present a problem 49 different times if the problem comes up in each of the 49 States.

Senator ANDERSON. Isn't that burdensome?

Mr. SPORN. It is not burdensome to the utility that is operating in only one of the States.

Senator ANDERSON. When you have radioactive materials going into streams or going into the skies, in the case of an incident, you do more than touch a single State?

Mr. SPORN. Yes.

Senator ANDERSON. You hardly use high pressure steam as being comparable to the problems that arise in connection with atomic

reactors because the threat is a little more widespread. It is not just a local threat.

Mr. SPORN. I think that is true. I also think and I later say so, that where there are interestate problems involved, I think in many cases those can be handled by bringing together three or four or five States involved. We have that example in the pollution problem in the Ohio River.

Senator ANDERSON. We have a problem right now in connection with Switzerland and Austria, where the Swiss are perfectly satisfied because if there is any incident, the fallout won't come on them. It will come on the Austrians. But the Austrians don't take it quite so calmly.

Mr. SPORN. That is right.

Senator ANDERSON. It may be that if you located a reactor against the adjoining State, State No. 1 at which it is located says it is all right. If it blows up, somebody else gets it. There is a slightly different case to be made for Federal regulation in the field of atomic reactors than in the field of the steam boilers.

Mr. SPORN. The case is different, but only in degree. I would like to say this in general, Senator. For many years, we operated in nine States of the United States. For the last 25 years or so we have very actively developed our system as a single system in 7 States, all contiguous. Many of the problems that we have affect more than one State. It has been our experience that some of the problems that affect two States are much better in the sense that they are more easily worked out, and we can work them out better at a State level. There are problems that cannot be worked out at the State level and in our operation we have had the problem of working these out, but under two major Federal agencies, the Federal Power Commission, and the Securities and Exchange Commission. In my judgment, looking ahead toward the future of this great country of ours, where certainly at the present time there are clear indications that in a relatively short time this will be a Nation of 250 million, maybe 50 States—surely 50 States, at least—that any of the problems that can be solved at the State level had better be solved at the State level. Any of the problems that affect more than one State ought to be solved to the extent that they can be solved by getting together the respective States. Where they are not soluble on that basis, they should not be a sort of Texas League area.

Senator ANDERSON. You have not been reading any of Governor Faubus' speeches lately.

Mr. SPORN. No, I have not.

Senator ANDERSON. There are different degrees to different problems. That is all I was trying to suggest, Mr. Sporn. I do believe we get along very well with State regulations in many instances.

Mr. SPORN. Yes, sir.

Senator ANDERSON. But where you have a device of this nature, I just wonder, for example, even though there is considerable development of atomic energy in my own State of New Mexico, how we could get an inspector on our State payroll that would be qualified to pass on a program for the construction of a great reactor, whereas the Atomic Energy Commission may have a man available. From that standard, I wonder if there is not some slight difference in the program.

Mr. SPORN. There is a difference. I am talking about the basic policy. I hope you did not take anything I have said here as a detailed outline for a program of legislation. I was discussing the basic philosophy. As to that, I have given a great deal of thought, and I have convictions which I hope will have some value.

Senator ANDERSON. I did not try to do that. As I have many times said to you, I believe you have done a very fine job with OVEC and your own private business. Because of that people will listen probably more to what you have to say than if you were a failure in business. I would hope so, at least.

Mr. SPORN. I would, too.

Senator ANDERSON. Therefore, I would hate to get you started off on the path that when it comes to regulating safety on these atomic devices, it must be done by the State. We are going to have some hearings on that. We have had this problem. The States have this problem. I am just hopeful that you reserve your judgment along with the rest of us.

Mr. SPORN. I have also indicated, Senator, that there could be different views and I am not one to say that these are unsound views. I give you my best conviction. Our experience therefore leads me to the judgment that the existing State agencies are the agencies to be given the responsibility for passing upon the safety and adequacy of programs of disposal worked out under State laws with the help of such Federal guides—and I would not omit from that agencies—as may be needed, and which will surely be forthcoming. It is true, of course, that the situation is not clearly visualizeable at the present time and may develop here which will have a strong interstate character and which will disqualify as to jurisdiction any one of the two or more States that might be involved in the matter. But as has been observed in many other cases of the same kind, in irrigation, in pollution, and water withdrawal, the State compact is an ideal instrument for that purpose, and can be used here. It has its advantages in that it brings together to the solution of a problem affecting more than one State the very States that are most vitally affected, and this element of directness of interest, it seems to me, is a strong argument for keeping out unnecessary Federal jursdiction or control as long as interstate involvements do not result in failure or even lessening of responsibility.

In making these final observations, I recognize, of course, that State and local agencies may need assistance from the Federal Government, with its initial mass of experience in the field to assure that State agencies acquire the benefit of such experience and develop their own qualified staffs to carry on the work. My hope is that this can be accomplished by cooperative arrangements between the AEC and the other Federal and State agencies, through temporary loans of personnel, grant-in-aid measures, and the like.

We have had much useful experience in this kind of cooperation in other fields. In the long run I am convinced that this will give us the best results in atomic energy, far better than a system of comprehensive permanent Federal regulation. Here, too, what I have stated represents an objective which cannot be obtained overnight, but rather must be achieved in an evolutionary process as the staffs of the

appropriate State agencies acquire the education and training necessary to enable them to assume their responsibilities.

In conclusion, I cannot emphasize too strongly that in this brief statement I have simply attempted to give expression to the principles which I believe should govern our approach to the problem of waste disposal from the industrial standpoint. It is necessary and possible to elaborate at great length on each of the points which I have touched. Such elaboration will occur in part in the course of these hearings. Much of it should take place subsequent to these hearings under the stimulus that these hearings provide. For my own part, if the committee would like to pursue any further matters with me, I am anxious to be helpful in any way I can.

Representative HOLIFIELD. Thank you, Mr. Sporn.

Mr. SPORN. Thank you, Mr. Chairman.

Representative HOLIFIELD. Are there any questions?

Mr. RAMEY. Do you think that we have been entirely successful in handling stream pollution?

Mr. SPORN. No, I don't think we have. On the other hand, I don't think we were working at the problem from the very beginning as we are working on this. I think the danger here is recognized much more clearly, and I would hope and believe more extensively. I think there was a period there when the possibility of pollution really was not given much thought in the case of streams. I think that possibility is recognized now. I think it is recognition that has brought about the major change in assuring the solution of the problem. I think it is fortunate that in the atomic case we do recognize the problem at many levels, and particularly right here in the Joint Committee, and I believe in the Atomic Energy Commission, also.

Representative HOLIFIELD. Thank you very much, sir.

Mr. SPORN. Thank you, Mr. Chairman?

Representative HOLIFIELD. Before we adjourn, I have several papers on "The Industrial Viewpoint Toward Radioactive Waste Disposal" for the record.

(The papers referred to follow:)

Submitted by Vice Adm. George F. Hussey Jr.

Born - June 15, 1894 in Brookline, Massachusetts

Admiral Hussey became Managing Director of the American Standards Association on January 1, 1948.

Having graduated from the U. S. Naval Academy with distinction in 1916, Admiral Hussey served on the USS PENNSYLVANIA throughout World War I. In 1920 he took postgraduate work in ordnance engineering at Annapolis, and the University of Chicago. During nearly thirty-six years of naval service he held many important fleet assignments. On December 10, 1941, Admiral Hussey was assigned to command the off-shore patrol of the Fourteenth Naval District in the Hawaiian Islands. His shore duty was associated with various sections of the Bureau of Ordnance of which he became Chief in December 1943, serving until September 1947. For this latter service he was awarded the Navy Distinguished Service Medal and was made an Honorary Commander of the Order of the British Empire. He was retired for physical disability on December 1, 1947.

In addition to his responsibilities as Managing Director and Secretary of the American Standards Association, Admiral Hussey is Vice President of the International Organization for Standardization. He is a Director of the American Ordnance Association; past president of the American Ordnance Association and past president of its New York Post; past president of the U. S. Naval Academy Alumni Association and past president of its New York Chapter. He is a fellow of the Standards Engineers Society.

The American Standards Association, which is the national clearinghouse for standards, is a federation of 64 technical societies, trade associations and consumer groups with 56 similar groups as Associate Members. It draws its support primarily from its more than 2,000 Company Members.

The ASA had its beginnings in 1918 when five engineering societies formed the American Engineering Standards Committee. These societies were The American Society of Mechanical Engineers, the American Institute of Electrical Engineers, the American Institute of Mining and Metallurgical Engineers, the American Society of Civil Engineers and the American Society for Testing Materials. They established the AESC for the purpose of eliminating conflicts and duplication among the standards which they themselves were issuing, each being authoritative in its own field. The first act of the AESC was to invite three government departments to join on an equal footing with the founding members. The War Department, the Navy Department and the Department of Commerce accepted. These eight Member-Bodies through their representatives on the Main Committee of the AESC developed the principles and procedures which governed the operations of the AESC and which basically govern today the operations of the ASA.

These principles are:

First, standardization and all of its aspects are voluntary actions.

Second, in the development of a standard any group having a substantial interest has an inherent right to participate in the proceedings.

Third, if a committee be organized for the development of standards, it must be so balanced among producers, consumers, distributors, general interests and regulatory bodies, if they be involved, so that no one group may have a preponderance. Thus no group by sheer weight of votes can force a standard through or conversely block a standard.

Fourth, to receive approval as American Standard a standard must be supported by a consensus of those substantially concerned with its scope and provisions. The loose word "consensus" was chosen advisedly because the ASA reviewing boards both count and weigh the votes. A negative vote within a committee must be accompanied by supporting reasons. There have been occasions when a single negative vote from a source importantly affected and supported by strong reasons has been sufficient to return the standard to the committee for further work. On other occasions a number of negative votes with comparatively trivial supporting reasons have been noted for consideration at the time of revision of the standard, and the standard has been approved to make it available to those who want it.

These principles still govern the work of the ASA. The consensus principle applies with equal force whether a standard is developed by a committee specially organized

or whether it comes from one of the large number of highly competent technical societies and trade associations which develop standards in their own fields. Any such standard may be submitted for approval as American Standard and will receive that approval if it meets the criteria of the ASA principles.

In 1928, to cope with the increased workload, the American Engineering Standards Committee was reorganized as the American Standards Association. By 1948, the operation involved half a million dollars a year and the Member-Bodies instructed the directors to incorporate under the laws of any state in order to secure corporate law protection and at the same time to continue to seek a Federal Charter. At this time there were ten Member-Bodies which were departments and agencies of the government. They were: Departments of Agriculture, Commerce, Interior, Labor, Navy, War, and Treasury, Government Printing Office, Federal Works Agency, and the Housing and Home Finance Agency. When the ASA became the American Standards Association, Incorporated under the membership corporation laws of the State of New York, the ten government Member-Bodies resigned on the grounds that the sovereign United States could not belong to a state-incorporated organization.

The present organization of the American Standards Association is shown on chart No. 1, attached.

The Standards Council is the direct descendant of the old Main Committee of the AESC and has responsibility for technical policies and the procedures for the application of the ASA principles. The Board of Directors is concerned with general policies, administration and fiscal matters. All Member-Bodies are represented on the Council and 15 at a time are represented on the Board of Directors. The detailed work of the Council is delegated to the standards boards which are authoritative in their fields. The final review of standards before approval is handled by the Board of Review - a six-man group elected by the Council from its own membership.

On June 7, 1955, The American Society of Mechanical Engineers sent the ASA a request "that ASA initiate a project on the safety codes for nuclear reactors, accelerators and other related subjects and consider The American Society of Mechanical Engineers for sponsorship." This request was shortly followed by similar requests from the Atomic Industrial Forum (September 13, 1955) and the National Electrical Manufacturers Association (September 14, 1955). In response to these requests the ASA called a General Conference which was held in Washington on December 8, 1955. The Conference was attended by 175 representatives of 119 organizations. It decided that the ASA should be requested to undertake standardization in the field of nuclear energy and requested "the ASA to appoint a Planning Committee to urgently study the question of standardization in the field of nuclear energy in detail and report back to the Conference as soon as possible." The Planning Committee was organized under the chairmanship of Mr. Morehead Patterson, Chairman of the Board of American Machine and Foundry Company. The Planning Committee recommended the establishment of a Nuclear Standards Board and, under that Board, the establishment of six sectional committees to deal with standardization in particular branches of the nuclear energy field. The organizations which had attended the Conference approved the recommendations of the Planning Committee which were in turn approved by the Standards Council.

The ASA's organization in the field of nuclear energy is shown on chart No. 2. The Nuclear Standards Board elected Mr. Patterson as its chairman and proceeded to establish the several sectional committees indicated. The Board meets from time to

time to receive reports of progress and to settle policy questions on behalf of the Standards Council within its own field.

The organization of the several sectional committees is shown on a series of charts Nos. 3 through 8.

The matter of disposal of radioactive industrial wastes is the responsibility of Subcommittee 2 of Committee N5. The organization of this subcommittee is shown on Chart No. 9. It is not required that subcommittees be balanced and it is, therefore, possible to concentrate on any subcommittee the best technical experts available in the field of the subcommittee. The protection of all interests through balanced membership is obtained in the parent sectional committee. The details of the work and the progress made so far are shown in the status report (CN 34), page 4, and the later report (CN 40), page 5, attached.

It should be emphasized that in this work the ASA as a federation and the ASA staff have no hand in the technical decisions. These are made by the experts drawn from industry, government and the scientific and technical communities who do their work at the expense and on the time of their employers. The ASA serves as the machinery through which these experts can come together and through which their conclusions can be examined on three scores:

First, does the proposed standard conflict with any standard already approved by the ASA?

Second, was the sectional committee which recommended the standard competent and properly organized? and

Third, is the proposed standard supported by a consensus?

The attention of the Committee is invited to the broad range of organizational participation in the Nuclear Standards Board as well as in the several sectional committees. It will be noted that there is extensive and active participation on the part of a considerable number of Federal Government activities, including particularly the Atomic Energy Commission, the National Bureau of Standards, and the Department of Defense.

Prepared by staff of the American Standards Association, Incorporated

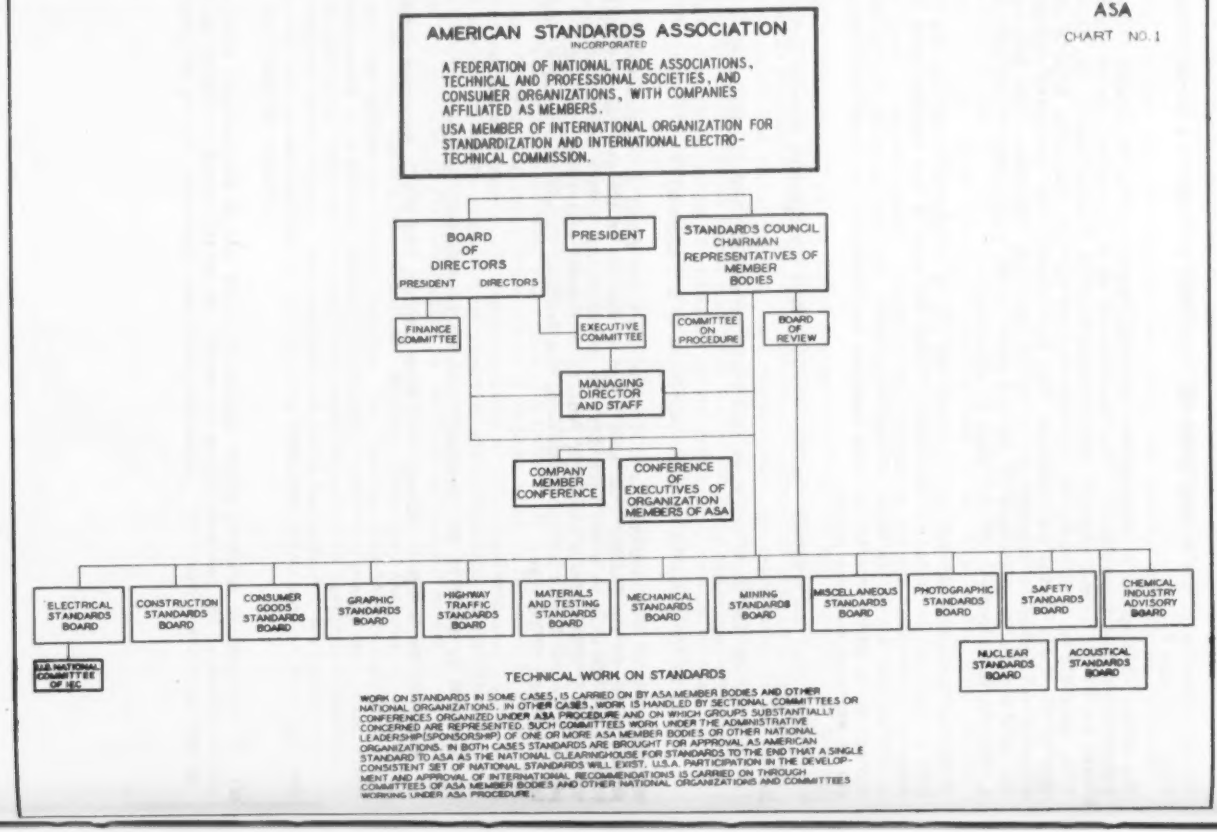
Submitted by George F. Hussey, Jr., Vice Admiral, USN (Ret), Managing Director

ASA
CHART NO. 1

AMERICAN STANDARDS ASSOCIATION
INCORPORATED

A FEDERATION OF NATIONAL TRADE ASSOCIATIONS, TECHNICAL AND PROFESSIONAL SOCIETIES, AND CONSUMER ORGANIZATIONS, WITH COMPANIES AFFILIATED AS MEMBERS.

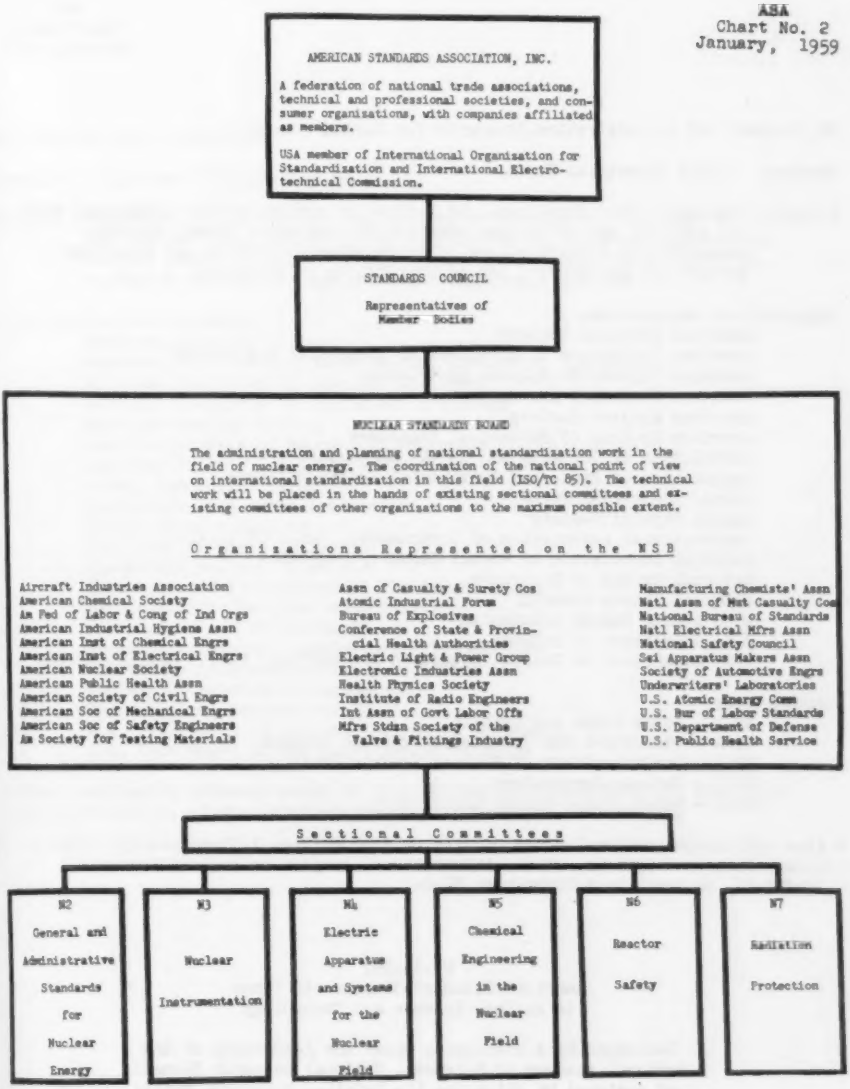
USA MEMBER OF INTERNATIONAL ORGANIZATION FOR STANDARDIZATION AND INTERNATIONAL ELECTRO-TECHNICAL COMMISSION.



TECHNICAL WORK ON STANDARDS

WORK ON STANDARDS IN SOME CASES, IS CARRIED ON BY ASA MEMBER BODIES AND OTHER NATIONAL ORGANIZATIONS. IN OTHER CASES, WORK IS HANDLED BY SECTIONAL COMMITTEES OR CONFERENCES ORGANIZED UNDER ASA PROCEDURE AND ON WHICH GROUPS SUBSTANTIALLY CONCERNED ARE REPRESENTED. SUCH COMMITTEES WORK UNDER THE ADMINISTRATIVE LEADERSHIP(SPONSORSHIP) OF ONE OR MORE ASA MEMBER BODIES OR OTHER NATIONAL ORGANIZATIONS. IN BOTH CASES STANDARDS ARE BROUGHT FOR APPROVAL AS AMERICAN STANDARD TO ASA AS THE NATIONAL CLEARINGHOUSE FOR STANDARDS TO THE END THAT A SINGLE CONSISTENT SET OF NATIONAL STANDARDS WILL EXIST. U.S.A. PARTICIPATION IN THE DEVELOPMENT AND APPROVAL OF INTERNATIONAL RECOMMENDATIONS IS CARRIED ON THROUGH COMMITTEES OF ASA MEMBER BODIES AND OTHER NATIONAL ORGANIZATIONS AND COMMITTEES WORKING UNDER ASA PROCEDURE.

ASA
Chart No. 2
January, 1959


AMERICAN STANDARDS ASSOCIATION, INC.
A federation of national trade associations, technical and professional societies, and consumer organizations, with companies affiliated as members.
USA member of International Organization for Standardization and International Electrotechnical Commission.
STANDARDS COUNCIL
Representatives of Member Bodies
NUCLEAR STANDARDS BOARD
The administration and planning of national standardization work in the field of nuclear energy. The coordination of the national point of view on international standardization in this field (ISO/TC 85). The technical work will be placed in the hands of existing sectional committees and existing committees of other organizations to the maximum possible extent.
Organizations Represented on the NSB
Aircraft Industries Association
American Chemical Society
Am Fed of Labor & Cong of Ind Orgs
American Industrial Hygiene Assn
American Inst of Chemical Engrs
American Inst of Electrical Engrs
American Nuclear Society
American Public Health Assn
American Society of Civil Engrs
American Soc of Mechanical Engrs
American Soc of Safety Engineers
Am Society for Testing Materials
Assn of Casualty & Surety Cos
Atomic Industrial Forum
Bureau of Explosives
Conference of State & Provincial Health Authorities
Electric Light & Power Group
Electronic Industries Assn
Health Physics Society
Institute of Radio Engineers
Int Assn of Govt Labor Offs
Mfrs Stdrn Society of the Valve & Fittings Industry
Manufacturing Chemists' Assn
Natl Assn of Mut Casualty Cos
National Bureau of Standards
Natl Electrical Mfrs Assn
National Safety Council
Sci Apparatus Makers Assn
Society of Automotive Engrs
Underwriters' Laboratories
U.S. Atomic Energy Comm
U.S. Bur of Labor Standards
U.S. Department of Defense
U.S. Public Health Service
Sectional Committees
N2 General and Administrative Standards for Nuclear Energy
N3 Nuclear Instrumentation
N4 Electric Apparatus and Systems for the Nuclear Field
N5 Chemical Engineering in the Nuclear Field
N6 Reactor Safety
N7 Radiation Protection

ASA
Chart No. 3
January, 1959

N2, General and Administrative Standards for Nuclear Energy

Sponsor: Atomic Industrial Forum

Scope*: Standards, specifications, and methods of administratior associated with the peaceful use of nuclear energy including color codes, symbols, nomenclature, qualifications of professionals, records and reporting systems and procedures, and accountability of materials.

Organizations Represented:
American Chemical Society
American Conference of Governmental Industrial Hygienists
American Industrial Hygiene Association
American Municipal Association
American Nuclear Society
American Society of Mechanical Engineers
Association of Casualty and Surety Companies
Association of State and Territorial Health Officers
Atomic Industrial Forum
Health Physics Society
International Association of Governmental Labor Officials
National Association of Mutual Casualty Companies
National Bureau of Standards
National Safety Council
U.S. Atomic Energy Commission
U.S. Department of Defense
U.S. Department of Health, Education and Welfare, Public Health Service

Subcommittees:
N2-1 - Color Codes and Symbols
N2-2 - Procedures for Industrial Exposure Records
N2-3 - Qualifications of Nuclear Professionals
N2-4 - Nuclear Terminology
N2-5 - Model Atomic Energy Legislation

* Also coordinates national point of view on Subcommittee 1, Terminology, Definitions, Units and Symbols, of the international project on Nuclear Energy, ISO/TC 85, through its subcommittee N2-4.

N1.1-1957
American Standard Glossary of Terms
in Nuclear Science and Technology

Developed by a conference under the leadership of the National Academy of Sciences, National Research Council, and approved by ASA under the Existing Standards Method.

ASA
Chart No. 4
January, 1959

N3, Nuclear Instrumentation

Sponsor: Institute of Radio Engineers

Scope*: Standards, specifications and methods of testing for instrumentation in the nuclear field including instrumentation for personnel protection, reactor control, industrial processes, analysis and laboratory work, radiation calibration equipment and components therefor.

Organisations Represented:
American Chemical Society
American Conference of Governmental Industrial Hygienists
American Institute of Chemical Engineers
American Institute of Electrical Engineers
American Nuclear Society
American Society of Mechanical Engineers
American Society for Testing Materials
Atomic Industrial Forum
Electronic Industries Association
Health Physics Society
Institute of Radio Engineers
Instrument Society of America
International Brotherhood of Electrical Workers
Manufacturing Chemists' Association
National Bureau of Standards
National Electrical Manufacturers Association
Scientific Apparatus Makers Association
Society of Automotive Engineers
Underwriters' Laboratories
U.S. Department of the Army, Corps of Engineers
U.S. Department of the Navy, Bureau of Ships

* Also coordinates national point of view on Technical Committee 45, Electrical Instrumentation, of the International Electrotechnical Commission.

ASA
Chart No. 5
January, 1959

N4, Electric Apparatus and Systems for the Nuclear Field

Sponsors: American Institute of Electrical Engineers
Electric Light and Power Group*
National Electrical Manufacturers Association

Scope: Standards for electrical power apparatus, systems and controls, used in the nuclear field.

Organizations Represented:
American Institute of Electrical Engineers
American Nuclear Society
American Society of Mechanical Engineers
Electric Light and Power Group
Electronic Industries Association
Elliot Company, Division of Carrier Corporation
General Electric Company
International Brotherhood of Electrical Workers
National Bureau of Standards
National Electrical Manufacturers Association
Underwriters' Laboratories
U.S. Department of the Army

* Edison Electric Institute and Association of Edison Illuminating Companies

ASA
Chart No. 6
January, 1959

N5, Chemical Engineering for the Nuclear Field

Sponsor: American Institute of Chemical Engineers

Scope*: Standards, specifications, tolerances, and methods of testing on the chemical engineering aspects of the nuclear field including: the refining, processing, separation, purification, treatment, packaging, handling, and disposal of fuels and allied materials such as radioisotopes, industrial chemicals, pharmaceuticals with isotope tracers, and radioactive wastes (gases, liquids, and solids); the use of radioactive sources for tracer purposes and for processing of foods and other materials; and the application and use of chemically resistant coatings and the cleaning of contaminated equipment and facilities.

Organizations Represented:
American Chemical Society
American Conference of Governmental Industrial Hygienists
American Industrial Hygiene Association
American Institute of Chemical Engineers
American Nuclear Society
American Public Health Association
American Society of Civil Engineers
American Society of Mechanical Engineers
American Society for Testing Materials
Association of Casualty and Surety Companies
Electric Light and Power Group
Health Physics Society
Mallinckrodt Chemical Works
Manufacturing Chemists' Association
National Association of Mutual Casualty Companies
National Board of Fire Underwriters
Scientific Apparatus Makers Association
Society of Automotive Engineers
Underwriters' Laboratories
U.S. Atomic Energy Commission
U.S. Department of Defense
U.S. Department of Health, Education and Welfare, Public Health Service

Subcommittees:
N5-1 - Fuel Manufacture and Fabrication
N5-2 - Radioactive Waste Disposal
N5-3 - Recovery of Irradiated Fuel
N5-4 - Use and Handling of Radioisotopes and High Energy Radiation
N5-5 - Packaging and Transportation of Radioactive Material

* Also coordinates national point of view on Subcommittee 4, Radioisotopes, of the international project on Nuclear Energy, ISO/TC 85, through its subcommittee N5-4.

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ASA
Chart No. 7
January, 1959

N6, Reactor Safety

Sponsors: American Nuclear Society
American Society of Mechanical Engineers

Scope*: Codes and standards concerned with the hazards involved in the design, location, construction, and operation of nuclear reactors and of potential critical assemblies.

Organizations Represented:

American Chemical Society
American Conference of Governmental Industrial Hygienists
American Federation of Labor and Congress of Industrial Organizations
American Hospital Association
American Industrial Hygiene Association
American Institute of Chemical Engineers
American Institute of Electrical Engineers
American Institute of Mining, Metallurgical and Petroleum Engineers
American Institute of Physics
American Iron and Steel Institute
American Medical Association
American Nuclear Society
American Public Health Association
American Society of Civil Engineers
American Society of Mechanical Engineers
American Society for Testing Materials
Associated General Contractors of America
Association of Casualty and Surety Companies
Atomic Industrial Forum
Compressed Gas Association
Electric Light and Power Group
Health Physics Society
Institute of Radio Engineers
Instrument Society of America
International Association of Machinists
International Brotherhood of Electrical Workers
Manufacturers Standardization Society of the Valve and Fittings Industry
Manufacturing Chemists' Association
Mechanical Contractors Association of America
National Association of Mutual Casualty Companies
National Association of Plumbing Contractors
National Board of Fire Underwriters
National Bureau of Standards
National Clay Pipe Manufacturers Association
National Electrical Manufacturers Association
National Industrial Zoning Committee
National Research Council
Scientific Apparatus Makers Association
Society of Automotive Engineers
Society of Naval Architects and Marine Engineers
Underwriters' Laboratories
U.S. Atomic Energy Commission
U.S. Department of Health, Education and Welfare, Public Health Service

N6, Reactor Safety (continued)

Subcommittees:

- N6-1 - Site Evaluation
- N6-2 - Containment
- N6-3 - Fluid Systems of the Reactor and Fuel Within the Reactor
- N6-4 - Reactor Dynamics and Control Requirements
- N6-5 - Instrumentation and Execution of Control Requirements
- N6-6 - Operation, Operator Qualifications, Inspection and Maintenance, and Records
- N6-7 - Failure Probabilities and Maximum Credible Accidents
- N6-8 - Fissionable Material Outside Reactors (normally subcritical systems)

* Also coordinates national point of view on Subcommittee 3, Reactor Safety, of the international project on Nuclear Energy, ISO/TC 85.

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ASA
Chart No. 8
January, 1959

N7, Radiation Protection

Sponsors: Atomic Industrial Forum
National Safety Council

Scope*: Safety standards for the protection of persons employed in facilities associated with the production and utilization of fissionable materials against the normal, routine hazards present in such facilities (facilities include mines, mills, refineries, separation plants, fuel element fabrication plants, critical assembly facilities, processing and reprocessing plants, working areas around nuclear reactors of all types, and transportation, and purification of "spent fuel elements" and gross fission products).

Organizations Represented:
American Chemical Society
American Federation of Labor and Congress of Industrial Organizations
American Industrial Hygiene Association
American Nuclear Society
American Public Health Association
American Society of Mechanical Engineers
American Society of Safety Engineers
Association of Casualty and Surety Companies
Association of State and Territorial Health Officers
Atomic Industrial Forum
Health Physics Society
International Association of Governmental Labor Officials
International Brotherhood of Electrical Workers
Manufacturing Chemists' Association
National Association of Mutual Casualty Companies
National Bureau of Standards
National Safety Council
Society of Automotive Engineers
Underwriters' Laboratories
U.S. Atomic Energy Commission
U.S. Department of Health, Education and Welfare, Public Health Service
U.S. Department of Labor, Bureau of Labor Standards

Subcommittees:
N7-1 - Uranium Mines and Mills
N7-2 - Uranium and Thorium Refineries
N7-3 - Isotopic Separation Plants
N7-4 - Health Physics of Fuel Element Fabrication
N7-5 - Health Physics for Reactors

* Also coordinates national point of view on Subcommittee 2, Radiation Protection, of the international project on Nuclear Energy, ISO/TC 85.

ASA
Chart No. 9
January, 1959

N5, Chemical Engineering for the Nuclear Field

Subcommittee 2 - Radioactive Waste Disposal

Chairman: Dr. J.A. Lieberman

Objective: Standards, codes, specifications, tolerances, and methods of testing on the chemical engineering aspects of the nuclear field concerning the handling, storage, treatment, processing, disposal and/or shipment of radioactive wastes.

Program of work:

1. Classification of Wastes (gaseous, liquid, and solid) - to include chemical and physical characteristics, degree of radioactivity, etc. Definitions of terminology (high- and low-level) uniform measurements, etc.

2. Permissible Waste Effluent Standards - evaluation of existing MPC's to take into account such environmental factors as dilution, downstream uses, sanitary and storm sewer systems, etc.

3. Packaging and Transportation of Wastes - to include container specifications, shielding requirements, quantities and types of material to be shipped, and possibly an analysis of transportation routes involved.

4. Standard Methods of Liquid Waste Treatment - for low and intermediate level wastes, based on activity level, radionuclide composition, and degree of treatment required. Description of various waste processing methods, decontamination factors to be expected, etc.

5. Standardization of Effluent Monitoring Procedures - pertaining to stack and liquid effluents. Uniformity in types and frequencies of samples to be collected, locations of sample stations, methods of analysis, counting techniques, etc., are items to consider.

6. Site Selection Criteria - pertaining to location and operation of all nuclear facilities from the standpoint of waste handling, processing, and storage considerations. Areas of consideration would include meteorology, geology, hydrology, seismology, topography, population data and surrounding activities.

7. Uniform Reporting System - need for specific standardized reporting procedures on quantities of radioactivity discharged to the environment in cases of accident and also for normal operation. Will aid in correlation of data from one location to another.

AMERICAN STANDARDS ASSOCIATION
INCORPORATED
70 East Forty-fifth Street New York 17, N. Y.
ASA

CN 40
November 17, 195

To the Members of Standards Council

Status Report of Projects Under the Jurisdiction of the Nuclear Standards Board

Gentlemen:

In accordance with Section 105 of ASA Procedure which requires that the ASA be kep informed of the progress of work, the status of the projects under the jurisdictio of the NSB is reported herein. All projects listed are under the Sectional Committee Method unless otherwise noted.

N1, Glossary of Terms in Nuclear Science and Technology
(Existing Standards Method)

N1.1-1957, American Standard Glossary of Terms in Nuclear Science and Technology

This standard was developed by a conference under the leadership of the National Academy of Sciences, National Research Council, and approve by ASA under the Existing Standards Method. The National Research Council has now assigned the copyright on this document to the American Standards Association, and the revision of this standard has been assigned to the N2 Sectional Committee.

N2, General and Administrative Standards for Nuclear Energy

Sponsor: Atomic Industrial Forum
Chairman: William A. McAdams
Secretary: Saul J. Harris

Scope: Standards, specifications, and methods of administration associated with the peaceful use of nuclear energy including color codes, symbols, nomenclature, qualifications of professionals, records and reporting systems and procedures, and accountability of materials.

Subcommittees:

N2-1, Color Codes and Symbols

Chairman: F. Raymond Zintz

This subcommittee, established to cover further the development of symbol proposals, has been working on a standard radiation symbol, but has met with opposition from the interstate transport agencies.

N2-2, Procedures for Industrial Exposure Records

Chairman: Arthur R. Keene

This subcommittee has been preparing a draft for a standard record keeping procedure.

N2 continued

N2-3, Qualifications of Nuclear Professionals

Chairman: Dr. G. Hoyt Whipple

This subcommittee has been working on standards for four types of professionals: Reactor Supervisor, Health Physicist, Nuclear Engineer, and Medical Radiation Specialist. There has been some problem created by the question of acceptance of the "nuclear engineer" as a separate specialty in engineering.

N2-4, Nuclear Terminology

Chairman: Jerome D. Luntz

This subcommittee met October 29, and started work on (a) revision of N1.1-1957, (b) international glossary (ISO/TC 85/SC 1), and (c) nuclear terms being considered by IEC/TC 1 documents 1(26)(Secretariat)265 and 1(66)(Secretariat)266.

N2-5, Model Atomic Energy Legislation

Chairman: Herbert Mermagen

This subcommittee, in the process of being organized, will have as its major purpose the review or development of model laws and codes for use by state and local governments in atomic energy generally.

The N2 Sectional Committee also coordinates the national point of view on Subcommittee 1, Terminology, Definitions, Units and Symbols, of the International project on Nuclear Energy, ISO/TC 85. (See Subcommittee N2-4).

The next meeting of the N2 Sectional Committee has been scheduled for December 4.

N3, Nuclear Instrumentation

Sponsor: Institute of Radio Engineers
Chairman: William H. Hamilton
Secretary: L.G. Cumming

Scope: Standards, specifications and methods of testing for instrumentation in the nuclear field including instrumentation for personnel protection, reactor control, industrial processes, analysis and laboratory work, radiation calibration equipment and components therefor.

The Committee to date has held three meetings: April 28, 1958, June 19, 1958 and September 25, 1958.

The April 28 meeting was mainly an organization meeting. The scope of work for the Committee was established as shown above. It was later submitted to the Nuclear Standards Board, and was approved by the NSB. At the organization

N3 continued

meeting it was recognized that there are many activities in standards work in the nuclear instrumentation field, and one of the biggest contributions of N3 would be to develop a coordinated statement showing all the activities and their inter-relationship. A survey was made of the work in all the professional societies associated with the Nuclear Standards Board by means of questionnaires, and from this survey an Index of Standards was developed. This lists the fields of interest of each of the technical societies which has worked in the field of nuclear instrumentation. It also lists the names and identification of standards which have been accepted. It is intended to keep this index up to date and publish it from time to time.

At the second meeting the Committee set up a subcommittee to handle the index mentioned above. A subcommittee on topical structure of standards was established to develop a pattern which can be used for the classification by topics of standards that are required in the nuclear instrument industry and in the general use of nuclear instrumentation. The Committee has agreed to a general policy of attempting to have standards developed by member organizations rather than writing these standards themselves.

At the third meeting some standards that had been prepared by the Electronic Industries Association were reviewed and comments made. Based on this review the standards will be rewritten and resubmitted at the time of the next Committee meeting. This represents the first real step in beginning to process standards that can be accepted for general use.

At the April 28 meeting the Chairman of the United States National Committee of the International Electrotechnical Commission discussed with the members of N3 a proposed IEC committee in the same general area of interest as that of N3. It was decided that N3 would cooperate to the fullest extent in the IEC work. At the September 25 meeting it was reported that IEC Technical Committee 45 (Electrical Measuring Instruments Used in the Application of Radioisotopes and Personal Protection) had been organized and that Germany had accepted the Secretariat. The German Secretariat had requested the United States to appoint a chairman for this committee. It was recommended by N3 that W.H. Hamilton be recommended as chairman of this committee. It was further recommended that R.F. Shea be appointed U.S. delegate to this committee.

The Committee hoped to hold its next meeting in New York City in December.

N4, Electrical Requirements for Reactors and Nuclear Power Systems and Generation and Application of Nuclear Radiation

Sponsors: American Institute of Electrical Engineers
Electric Light and Power Group
National Electrical Manufacturers Association
Chairman: Philip N. Ross
Secretary: Richard W. Shaul

Proposed scope: Standards for electrical power apparatus, systems and controls, used in the nuclear field.

N4 continued

The Sectional Committee held its first meeting on November 6, 1958 at which time it voted on a change in scope to that shown above (subject to the approval of the NSB) and a change in title to that of "Electric Apparatus and Systems for the Nuclear Field." Also at that meeting three planning subcommittees were established to study and recommend a subcommittee structure which would be practical in dealing with the scope of this project.

N5, Chemical Engineering for the Nuclear Field

Sponsor: American Institute of Chemical Engineers
Chairman: Stanley L Winde
Vice Chairman: Wheaton W. Kraft

(As of January 1, 1959, W.W. Kraft will become Chairman, and Stephen Lawroski will be Vice Chairman.)

Scope: Standards, specifications, tolerances, and methods of testing on the chemical engineering aspects of the nuclear field including: the refining, processing, separation, purification, treatment, packaging, handling, and disposal of fuels and allied materials such as radioisotopes, industrial chemicals, pharmaceuticals with isotope tracers, and radioactive wastes (gases, liquids, and solids); the use of radioactive sources for tracer purposes and for processing of foods and other materiels; and the application and use of chemically resistant coatings and the cleaning of contaminated equipment and facilities.

Organizations added since April 21: American Conference of Governmental Industrial Hygienists, National Board of Fire Underwriters, Underwriters' Laboratories, Inc.

A Sectional Committee meeting was held on March 12 at the ASA offices. Each subcommittee reported on the progress to date, and a fifth subcommittee was established on Packaging and Transportation of Radioactive Material. The programs outlined by the subcommittees are as follows:

Subcommittees:

N5-1, Fuel Manufacture and Fabrication

Chairman: Dr. C.E. Crompton

Dr. Crompton summarized two projects for this subcommittee:

1. Make an effort to come up with a standard scale on enrichment up to five percent which would be compatible with the type of reactor design considered. This could produce better service and lower cost, and

2. Solicit specifications that various producers and customers feel are appropriate for uranium oxide fuel.

N5 continued

N5-2, Radioactive Waste Disposal

Chairman: Dr. J.A. Lieberman

Dr. Lieberman has set up committees to determine which aspects of their scope can be standardized most profitably at this time. The committees are as follows:

Committee	Responsible	Assisting
1 - Classification of Wastes	R.J. Morton	E.G. Struxness I.B. Dick
2 - Permissible Waste Effluent Standards	E.G. Struxness	R.J. Morton F.W. Davis C.P. Straub
3 - Packaging and Transportation of Wastes	L.A. Barrer	J.A. Lieberman N.B. Tweed
4 - Standard Methods of Liquid Waste Treatment	F.W. Davis	I.B. Dick W.W. Kraft
5 - Standardization of Effluent Monitoring Procedures	C.P. Straub	J.W. Lake J.M. Smith
6 - Site Selection Criteria	J.M. Smith	N.B. Tweed L.A. Barrer E.G. Struxness
7 - Uniform Reporting System	J.W. Lake	C.P. Straub F.W. Davis

N5-3, Recovery of Irradiated Fuel

Chairman: Dr. C.E. Winters

It was the consensus of the group that Subcommittee 3 should continue to keep an eye on the situation and investigate the possibility of coordinating the activities that are in progress in the Atomic Energy Commission on the design of casks for elements.

N5-4, Use and Handling of Radioisotopes and High Energy Radiation

Acting Chairman: A.C. Jealous
Secretary: C.R. Russell

Because of a change in work assignment, H.J. Ogorzaly found it necessary to resign as chairman of this subcommittee.

A meeting of this subcommittee was held October 14 to elect a new chairman, and to consider the participation of this subcommittee in Subcommittee 4, Radioisotopes, of the International project on Nuclear Energy, ISO/TC 85.

Mr. A.C. Jealous agreed to accept the responsibility of Acting Chairman, and Dr. C.R. Russell of Secretary.

N5 continued

There was considerable discussion of the proposed outline of work of ISO/TC 85/SC 4 - Radioisotopes. While the entire scope, as proposed, is broader than the present scope of this subcommittee, it was agreed that N5-4 was the most logical group to accept this assignment. This had been the conclusion also of Mr. W.A. McAdams (Chairman of N2) and Mr. R.G. McAllister (Chairman of N7).

It was agreed to proceed to supplement the membership of this subcommittee in order to handle this assignment.

N5-5, Packaging and Transportation of Radioactive Material

Chairman: M.M. Braidech

Mr. Braidech has agreed to set up an organizational meeting of this subcommittee this fall to establish a written scope for the subcommittee, and to see what further committees should be organized.

The N5 Sectional Committee also coordinates the national point of view on Subcommittee 4, Radioisotopes, of the International project on Nuclear Energy, ISO/TC 85 (See Subcommittee N5-4).

N6, Reactor Safety Standards

Sponsors: American Nuclear Society
American Society of Mechanical Engineers
Chairman: Alfred Iddles
Vice Chairmen: Harold Etherington
Miles C. Leverett
Secretary: Melvin Ross

Scope: Codes and standards concerned with the hazards involved in the design, location, construction, and operation of nuclear reactors and of potential critical assemblies.

Subcommittees:

N6-1, Site Evaluation

Chairman: R.O. Brittan

N6-2, Containment

Chairman: Alf Kolflat

N6-3, Fluid Systems of the Reactor and Fuel Within the Reactor

Chairman: W.C. Gumprich

N6-4, Reactor Dynamics and Control Requirements

Chairman: John Ozeroff

N6 continued

N6-5, Instrumentation and Execution of Control Requirements

Chairman: John M. Finan

N6-6, Operation, Operator Qualifications, Inspection and Maintenance, and Records

Chairman: Charles F. Jones

N6-7, Failure Probabilities and Maximum Credible Accidents

Chairman: Dr. O.H. Greagor

N6-8, Fissionable Material Outside Reactors (normally subcritical systems)

Chairman: A.D. Callihan

A meeting of the N6 Steering Committee and chairmen of N6 subcommittees was held on July 15 at which time reports of subcommittees were heard. There was some discussion clarifying subject matter and scope of activity of various subcommittees. Committee memberships were reported virtually complete, and the writing of sections of final reports has begun. As a result of a coordination meeting held in Cincinnati on April 9 between various interests active in the nuclear standards field, it was reported that some progress has been made in unifying independent efforts in certain standards work.

Discussion was also held on the part codes and standards play in local and state legislation. Reference was made to the conference, sponsored by the Atomic Industrial Forum, to discuss this problem.

Consideration was also given to three American proposals which had been presented at the meeting of ISO/85 on Nuclear Energy (on whose Subcommittee 3, Reactor Safety, the N6 Sectional Committee coordinates the national point of view) in Harrogate, England, and the comments received at the Harrogate meeting. Copies of the American proposals were distributed to those attending for further study by the N6 subcommittees.

N7, Radiation Protection

Sponsors:	Atomic Industrial Forum National Safety Council
Chairman:	R.G. McAllister
Vice Chairman:	Hugh F. Henry
Secretary:	Saul J. Harris

Scope: Safety standards for the protection of persons employed in facilities associated with the production and utilization of fissionable materials against the normal, routine hazards present in such facilities (facilities include mines, mills, refineries, separation plants, fuel element fabrication plants, critical assembly facilities, processing and reprocessing plants, working areas around nuclear reactors of all types, and transportation, and purification of "spent fuel elements" and gross fission products).

N7 continued

Subcommittees:

N7-1, Uranium Mines and Mills

Chairman: Duncan Holaday

The chairman of this subcommittee wrote a preliminary rough draft for Radiation Protection Standards in Uranium Mines and Mills which he sent to the Chairman of the sectional committee who expanded it considerably. The next meeting of this subcommittee has been scheduled for November 17 in Denver, Colorado.

N7-2, Safety Standards in Uranium and Thorium Refineries

Chairman: Mont G. Mason

The chairman of this subcommittee believes that organizing the Symposium on Toxicity of Uranium put on by the AEC at the NYU Medical Center in October was a major contribution toward development of standards for uranium refineries.

N7-3, Isotopic Separations

Chairman: Dr. Hugh F. Henry

This subcommittee has been organized with the following membership:

R.C. Baker, Paducah Gaseous Diffusion Plant, UCNC
H.F. Henry, Oak Ridge Gaseous Diffusion Plant, UCNC
Ben Kalmon, Portsmouth Area Plant, GAT
J.D. McLendon, Y-12 Plant, UCNC

N7-4, Health Physics of Fuel Element Fabrication

Chairman: S.F. Audia

This subcommittee is still in the process of being organized. It planned to hold its second meeting on October 29 in Attleboro, Massachusetts.

N7-5, Health Physics for Reactors

Chairman: Joseph Fitzgerald

The first full meeting of this subcommittee was held October 14. The chairman has organized the work for the subcommittee, has drawn up an outline of the proposed standard, and has assigned sections to different committee members to write.

The N7 Sectional Committee held a meeting on October 14 for the purpose of receiving reports from the Subcommittee chairmen regarding progress made toward

N7 continued

drawing up Radiation Protection Standards, under their particular scopes.

The N7 Sectional Committee also coordinates the national point of view on Subcommittee 2, Radiation Protection, of the International project on Nuclear Energy, ISO/TC 85.

Henry G. Lamb, Secretary
Nuclear Standards Board

AMERICAN STANDARDS ASSOCIATION
INCORPORATED

ASA

CN 34
April 21, 1958

To the Members of Standards Council

Status Report of Projects Under the Jurisdiction of the Nuclear Standards Board

Gentlemen:

In accordance with Section 105 of ASA Procedure which requires that the ASA be kept informed of the progress of work, the status of the projects under the jurisdiction of the NSB is reported herein. All projects listed are under the Sectional Committee Method unless otherwise noted.

N1, Glossary of Terms in Nuclear Science and Technology
(Existing Standards Method)

N1.1-1957, American Standard Glossary of Terms in Nuclear Science and Technology

This standard was developed by a conference under the leadership of the National Academy of Sciences, National Research Council, and approved by ASA under the Existing Standards Method. The National Research Council has now assigned the copyright on this document to the American Standards Association, and the revision of this standard has been assigned to the N2 Sectional Committee.

N2, General and Administrative Standards for Nuclear Energy

Sponsor: Atomic Industrial Forum
Chairman: William A. McAdams
Secretary: Saul J. Harris

Scope: Standards, specifications, and methods of administration associated with the peaceful use of nuclear energy including color codes, symbols, nomenclature, qualifications of professionals, records and reporting systems and procedures, and accountability of materials.

Organizations Represented: American Chemical Society; American Industrial Hygiene Association; American Municipal Association; American Nuclear Society; American Society of Mechanical Engineers; Association of Casualty and Surety Companies; Association of State and Territorial Health Officers; Atomic Industrial Forum; Health Physics Society; International Association of Governmental Labor Officials; National Association of Mutual Casualty Companies; National Bureau of Standards; National Safety Council; U.S. Atomic Energy Commission; U.S. Department of Defense; U.S. Department of Health, Education and Welfare, Public Health Service.

Subcommittees:

N2.1, Color Codes and Symbols
This subcommittee has been working on a standard radiation symbol, but has met with opposition from the interstate transport agencies.

N2 continued

N2.2, Procedures for Industrial Exposure Records

Chairman: A.R. Keene

This subcommittee has met and organized its work, including scope. The members are now trying to draft a proposal for consideration among themselves. They hope to have a radiation recording system proposal for industry by the end of the current year.

N2.3, Qualifications of Nuclear Professionals

Chairman: Dr. G. Hoyt Whipple

This subcommittee has been organized, and is attempting to write qualification procedures for four types of professionals - (a) Nuclear Engineer (responsibility assigned to the American Nuclear Society which already has a procedure in operation), (b) Health Physicist (responsibility assigned to Health Physics Society), (c) Reactor Supervisor (responsibility assigned to Atomic Energy Commission), and (d) Reactor Supervisor (Medical) (responsibility assigned to various medical groups).

N2.4, Nuclear Terminology

Chairman: Jerome D. Luntz

This subcommittee has received formal authorization from the National Research Council to revise the Glossary of Terms in Nuclear Science and Technology, N1.1-1957. The members have been working on the Glossary as preparation for the meeting of ISO/TC 85/SC 1 in Harrogate in June. Their review of the terms has been for their own discussion purposes so far.

N2.5, Model Atomic Energy Legislation

This subcommittee has not yet been organized. On the agenda is the development of a series of model codes and laws to include radiation protection. Also to be considered is legislation on workmen's compensation.

The N2 Sectional Committee also coordinates the national point of view on Subcommittee 1, Terminology, Definitions, Units and Symbols, of the International Project on Nuclear Energy, ISO/TC 85.

N3, Nuclear Instruments

Sponsor: Institute of Radio Engineers
Chairman: William H. Hamilton
Secretary: L.G. Cumming

Proposed Scope: Standards, specifications and methods of testing for instruments used in the nuclear field including instruments for personnel protection, reactor control, industrial processes, analysis and laboratory work, radiation calibration equipment and components therefor.

N3 continued

Organizations Represented: American Chemical Society; American Institute of Electrical Engineers; American Nuclear Society; American Society of Mechanical Engineers; Atomic Industrial Forum; Electronic Industries Association; Health Physics Society; Institute of Radio Engineers; Instrument Society of America; International Brotherhood of Electrical Workers; Manufacturing Chemists' Association; National Bureau of Standards; National Electrical Manufacturers Association; Scientific Apparatus Makers Association; Society of Automotive Engineers; U.S. Department of the Navy, Bureau of Ships.

The first meeting of this sectional committee has been called for April 28.

N4, Electrical Requirements for Reactors and Nuclear Power Systems and Generation and Application of Nuclear Radiation

Sponsors: American Institute of Electrical Engineers
Electric Light and Power Group
National Electrical Manufacturers Association
Chairman: Philip N. Ross
Secretary: Richard W. Shaul

Proposed Scope: Standards, specifications and methods of testing for the electrical equipment used in nuclear reactors and nuclear power systems and in the generation and application of nuclear radiation.

N5, Chemical Engineering for the Nuclear Field

Sponsor: American Institute of Chemical Engineers
Chairman: Stanley I. Winde
Vice Chairman: Wheaton W. Kraft

Scope: Standards, specifications, tolerances, and methods of testing on the chemical engineering aspects of the nuclear field including: the refining, processing, separation, purification, treatment, packaging, handling, and disposal of fuels and allied materials such as radioisotopes, industrial chemicals, pharmaceuticals with isotope tracers, and radioactive wastes (gases, liquids, and solids); the use of radioactive sources for tracer purposes and for processing of foods and other materials; and the application and use of chemically resistant coatings and the cleaning of contaminated equipment and facilities.

Organizations Represented: American Chemical Society; American Industrial Hygiene Association; American Institute of Chemical Engineers; American Nuclear Society; American Public Health Association; American Society of Civil Engineers; American Society of Mechanical Engineers; American Society for Testing Materials; Association of Casualty and Surety Companies; Electric Light and Power Group; Health Physics Society; Manufacturing Chemists' Association; National Association of Mutual Casualty Companies; Scientific Apparatus Makers Association; Society of Automotive Engineers; U.S. Atomic Energy Commission; U.S. Department of Defense; U.S. Department of Health, Education and Welfare, Public Health Service.

N5 continued

An organizational meeting of the N5 Sectional Committee was held June 12, 1957 at the ASA offices. Twenty-six people attended, representing fifteen technical societies and interested groups. The following subcommittees were formed to study the problem in the various fields. (Each subcommittee has organized and agreed to a scope of work as shown):

N5.1, Fuel Manufacture and Fabrication

Chairman: Dr. C.E. Crompton

Scope: Feed materials processing and fuel element manufacture involving both normal and enriched nuclear material, but excluding the operations within the gaseous diffusion cascades.

Standards, codes, specifications, tolerances and methods of testing on the chemical engineering aspects of the nuclear field concerning:

1. The handling and processing of nuclear material during its residence at the concentrating mills.

2. The separations and purification steps leading ultimately to reactor grade oxide or metal.

3. The forming and fabrication of finished fuel elements and their subsequent inspection.

4. The unique nuclear aspects, such as criticality, of preparing fuel elements for shipment to the reactor sites.

5. The preparation of new fuel elements from decontaminated nuclear material resulting from the recycle of spent fuel.

N5.2, Radioactive Waste Disposal

Chairman: Dr. J.A. Lieberman

Scope: Standards, codes, specifications, tolerances, and methods of testing on the chemical engineering aspects of the nuclear field concerning the handling, storage, treatment, processing, disposal and/or shipment of radioactive wastes, and including:

1. Classification of Wastes (gaseous, liquid, and solid) - to include chemical and physical characteristics, degree of radioactivity, etc. Definitions of terminology (high and low-level) uniform measurements, etc.

2. Permissible Waste Effluent Standards - Evaluation of existing MPC's to take into account such environmental factors as dilution, downstream uses, sanitary and storm sewer systems, etc.

N5 continued

3. Packaging and Transportation of Wastes - To include container specifications, shielding requirements, quantities and types of material to be shipped, and possibly an analysis of transportation routes involved.

4. Standard Methods of Liquid Waste Treatment - For low and intermediate level wastes, based on activity level, radionuclide composition, and degree of treatment required. Description of various waste processing methods, decontamination factors to be expected, etc.

5. Standardization of Effluent Monitoring Procedures - Pertaining to stack and liquid effluents. Uniformity in types and frequencies of samples to be collected, locations of sample stations, methods of analysis, counting techniques, etc., are items to consider.

6. Site Selection Criteria - Pertaining to location and operation of all nuclear facilities from the standpoint of waste handling, processing, and storage considerations. Areas of consideration would include meteorology, geology, hydrology, seismology, topography, population data and surrounding activities.

7. Uniform Reporting System - Need for specific standardized reporting procedures on quantities of radioactivity discharged to the environment in cases of accident and also for normal operation. Will aid in correlation of data from one location to another.

N5.3, Recovery of Irradiated Fuel

Chairman: Dr. C.E. Winters

Scope: Standards, codes, specifications, tolerances and methods of testing on the chemical engineering aspects of the nuclear field concerning:

1. The transportation of used fuel assemblies from the reactor to the Irradiated Fuel Processing Plant (IFPP).

2. The reprocessing of the fuel elements in the IFPP.

3. The preparation for shipment of recovered fuels suitable for fuel fabrication activities, fob, IFPP.

4. The preparation for shipment of radioactive wastes suitable for waste disposal activities, fob, IFPP.

5. The preparation for shipment of fission product radioisotopes suitable for sale or other useful purposes.

6. The pre- and post-irradiation processing and the delivery of substances irradiated for the purposes of generating useful radioactivity, (non-fission radioisotope manufacture), fob reactor or isotope plant.

N5 continued

7. The application and use of chemical resistant coatings, cleaning facilities and decontamination of equipment.

N5.4, Use and Handling of Radioisotopes and High Energy Radiation

Chairman: H.J. Ogorzaly

Scope: Standards, codes, specifications, tolerances, and methods of testing on the chemical engineering aspects of the nuclear field concerning:

1. The use and handling of low intensity sealed radioisotope sources, e.g., container specifications, leak tests, handling of spills, disposal problems.

2. The use and handling of radiotracers, e.g., facilities and equipment needed, hygiene, and disposal.

3. Industrial use of radioisotopes, e.g., container specifications, handling, safety precautions.

4. High intensity radiation facilities, e.g., shielding and other safety precautions, source intensity calibrations, dosimetry.

The second sectional committee meeting was held March 12, 1958 at the ASA offices. Each subcommittee reported progress in its area of interest, and defined specific projects for further study. A fifth subcommittee was established -

N5.5, Packaging and Transportation of Radioactive Material

Chairman: M.M. Braidech

Anyone interested in this work or who has suggestions for projects in this field should contact the Chairman.

N6, Reactor Safety Standards

Sponsors: American Nuclear Society
American Society of Mechanical Engineers
Chairman: Alfred Iddles
Vice Chairmen: Harold Etherington
Miles C. Leverett
Secretary: Melvin Ross

Scope: Codes and standards concerned with the hazards involved in the design, location, construction, and operation of nuclear reactors and of potential critical assemblies.

Organizations Represented: American Chemical Society; American Conference of Governmental Industrial Hygienists; American Federation of Labor and Congress of Industrial Organizations; American Hospital Association; American Industrial Hygiene Association; American Institute of Chemical Engineers; American

N6 continued

Institute of Electrical Engineers; American Institute of Mining, Metallurgical and Petroleum Engineers; American Institute of Physics; American Iron and Steel Institute; American Medical Association; American Nuclear Society; American Society of Civil Engineers; American Society of Mechanical Engineers; American Society for Testing Materials; Associated General Contractors of America; Association of Casualty and Surety Companies; Atomic Industrial Forum; Compressed Gas Association; Electric Light and Power Group; Health Physics Society; Institute of Radio Engineers; Instrument Society of America; International Association of Machinists; International Brotherhood of Electrical Workers; Manufacturing Chemists' Association; Mechanical Contractors Association of America; National Association of Mutual Casualty Companies; National Association of Plumbing Contractors; National Board of Fire Underwriters; National Bureau of Standards; National Clay Pipe Manufacturers Association; National Electrical Manufacturers Association; National Industrial Zoning Committee; National Research Council; Scientific Apparatus Makers Association; Society of Automotive Engineers; Society of Naval Architects and Marine Engineers; U.S. Atomic Energy Commission; U.S. Department of Health, Education and Welfare, Public Health Service.

Subcommittees:

N6.1, Site Evaluation

Chairman: R.O. Brittan

Objective: To determine which criteria should be considered in the evaluation of the safety of possible reactor sites, to recommend methods of computation for site evaluation and to prepare, where possible, appropriate standards.

N6.2, Containment

Chairman: Alf Kolflat

Objective: This subcommittee shall, in those cases where containment is deemed necessary, determine the applicable standards.

N6.3, Fluid Systems of the Reactor and Fuel Within the Reactor

Chairman: W.C. Gumprich

Objective: Mechanical design, specifications, and pre-operational inspection and testing of the containment system of radioactive fluids of the reactor system. Will include non-radioactive fluid systems where possibility of hazard exists. Also, considerations peculiar to specific coolants, and the power supply and drives for pumps essential to safe operation of a reactor.

N6 continued

N6.4, Reactor Dynamics and Control Requirements

Chairman: John Ozeroff

Objective: Methods of predicting, evaluating and controlling against variations in reactor power, temperature or pressure which might lead to fission product release or endanger the reactor or plant equipment.

N6.5, Instrumentation and Execution of Control Requirements

Chairman: John M. Finan

Objective: To establish standard criteria for the quantity and kind of instruments, control operating mechanisms, and control panels, all as required to maintain the reactor, turbine generator (when used) and ancillary equipment in safe operation under all load conditions.

N6.6, Operation, Operator Qualifications, Inspection and Maintenance, and Records

Chairman: Charles F. Jones

Objective: To formulate standards or recommendations for operating procedures, operator qualifications, inspection - both prior to and subsequent to initial operation, maintenance procedures, and records required for safety

N6.7, Failure Probabilities and Maximum Credible Accidents

Chairman: W.R. Kanne

Objective: To establish guides of either acceptable probabilities or those which can be expected to obtain from failure frequency of reactor systems, and to define the concept of a "maximum credible accident" and establish recommended procedures for arriving at specific descriptions and magnitudes of maximum credible accidents.

N6.8, Fissionable Material Outside Reactors (normally subcritical systems)

Chairman: A.D. Callihan

Objective: To establish realistic standards (and guiding information) designed to prevent critical accumulation of fissionable material in operations, such as fuel element fabrication or fuel processing, fuel processing after use, recovery of excess fissionable material, attendant storage, and transportation.

The N6 Sectional Committee has held two meetings. It has also established a Steering Committee composed of the officers and the chairmen of the eight subcommittees. This Steering Committee has held three meetings, the last of which was March 19 in Chicago.

N6 continued

The N6 Sectional Committee also coordinates the national point of view on Subcommittee 3, Reactor Safety, of the International Project on Nuclear Energy, ISO/TC 85.

N7, Radiation Protection

Sponsors: Atomic Industrial Forum
National Safety Council
Chairman: Remus G. McAllister
Vice Chairman: Hugh F. Henry
Secretary: Saul J. Harris

Scope: Safety standards for the protection of persons employed in facilities associated with the production and utilization of fissionable materials against the normal, routine hazards present in such facilities (facilities include mines, mills, refineries, separation plants, fuel element fabrication plants, critical assembly facilities, processing and reprocessing plants, working areas around nuclear reactors of all types, and transportation, and purification of "spent fuel elements" and gross fission products).

Organizations Represented: American Association of State and Territorial Health Officers; American Chemical Society; American Federation of Labor and Congress of Industrial Organizations; American Industrial Hygiene Association; American Nuclear Society; American Public Health Association; American Society of Mechanical Engineers; American Society of Safety Engineers; Association of Casualty and Surety Companies; Atomic Industrial Forum; Health Physics Society; International Association of Governmental Labor Officials; Manufacturing Chemists' Association; National Association of Mutual Casualty Companies; National Bureau of Standards; National Safety Council; Society of Automotive Engineers; U.S. Atomic Energy Commission; U.S. Department of Health, Education and Welfare, Public Health Service; U.S. Department of Labor, Bureau of Labor Standards.

Subcommittees:

N7.1, Uranium Mines and Mills

Chairman: Duncan Holaday

This subcommittee has been organized. The U.S. Public Health Service is doing research on miners in uranium mines, and it is hoped that this research can be considered by the subcommittee.

N7.2, Safety Standards in Uranium and Thorium Refineries

Chairman: Mont Mason

This subcommittee is in the process of being organized.

N7 continued

N7.3, Isotopic Separations

Chairman: Dr. Hugh F. Henry

This subcommittee has not held a formal meeting, but through informal discussions it has been determined that there are three unclassified documents concerning health and safety of a gaseous diffusion plant which the subcommittee plans to consider.

N7.4, Health Physics of Fuel Element Fabrication

Chairman: James Harte

This subcommittee has not yet met.

N7.5, Health Physics for Reactors

Chairman: Joseph Fitzgerald

This subcommittee hopes to start work soon on the subject on transportation.

The sectional committee has held two meetings. The first, in September 1957, was for the purpose of organization and development of a scope. The second, in December 1957, produced some results. It was decided to go outside the membership of the sectional committee in order to obtain the technical knowledge to draw up health protection standards for uranium and thorium mines.

The N7 Sectional Committee also coordinates the national point of view on Subcommittee 2, Radiation Protection, of the International Project on Nuclear Energy, ISO/TC 85.

Henry G. Lamb

Henry G. Lamb, Secretary
Nuclear Standards Board

STATEMENT ON INDUSTRIAL PARTICIPATION IN WASTE DISPOSAL

PREPARED BY THE OFFICE OF THE ASSISTANT DIRECTOR FOR COMMERCIAL ACTIVITIES, DIVISION OF REACTOR DEVELOPMENT, U. S. ATOMIC ENERGY COMMISSION

Provision for the safe disposal of radioactive waste material is a necessity for the development of an atomic energy industry. The very real potential dangers associated with such waste and the public awareness thereof, have combined to make the need for this service on a completely safe basis practically a precondition for the existence of our industry.

The AEC is charged with the responsibility of enforcing such standards of safety in waste disposal activities as are appropriate for the protection of the public. While the exact standards and operating procedures will differ from operation to operation, depending upon the type and amount of waste involved, as well as other factors, there are two broad categories into which all waste disposal activities can be divided. The first category covers the disposal and custody of radioactive waste material of such nature and quantity that there exists a long term problem of custodial care and protection of the public. (For the purpose of this statement it is assumed that long term means a period of 10 years or more.) The second category covers waste disposal activities in which there is no such long term problem, and which do not require long term custodial care by the disposer.

This distinction between waste disposal operations which do and do not have a long term hazard associated with them and require custodial care by the disposer is particularly important from the point of view of industrial participation. The factors which must be considered in licensing a waste disposal operation differ greatly, depending upon the existence of a long term hazard. For instance, if a waste disposal operation does not involve a long term hazard, the AEC, can and does license the disposal activity

solely on the basis of such factors as nature of the site, method of packaging, disposal and the like. Examples of such waste disposal operations which belong in this category are sea disposal, as currently authorized, and the various small and non-commercial burial operations currently licensed under AEC Regulations, Part 20 and 30. Sea disposal is included in this category because, even though there is a potential long term hazard associated with it, the disposer will not be required to exercise any custodial care over the material which he has abandoned. Surveillance of sea disposal areas is a proper function of the Federal Government.

The situation is different for waste disposal operations which do involve a long term hazard and would require long term custodial care by the disposer. These would include most commercial waste burial operations on land. There the concern of AEC is not merely focused on the short term problem of packaging and disposal, but also on the long term problem of custody of the material and site, as a necessary measure for the protection of public health and safety. To guard against this long term hazard, steps must be taken for instance, to insure the continuity of custodial care of a burial site, to prevent abandonment, and perhaps even to provide for some financial protection if the hazard contained there is great enough to threaten injury to third parties.

These steps would present no problem if the burial site involved is a Government-owned facility, for the Government's operation would probably continue as long as the hazard exists and thus the protection of the public would be assured.

A problem would exist, however, if such an operation were privately owned and licensed, for here one cannot assume that custody would continue

as long as the hazard exists. The AEC, before licensing such an operation, could require some assurance that the property would not be abandoned, and that custody would be maintained for as long as the hazard exists. But giving such assurances would be no simple matter. The period for which custody of a facility will be required may be a hundred years or more, and to furnish a guaranty that the company involved would remain in existence for all this time and perform its job would raise all sorts of legal and administrative difficulties.

But assuming that these difficulties could be overcome and that all the assurances that could reasonably be expected are given, there would still be no certainty that private operation of a waste disposal facility would continue for all this time, the guaranties notwithstanding. First, there is a strong economic incentive for a company to pull up stakes as soon as all burial space is filled, because at that point the facility has really lost its economic usefulness. Second, there are many unforeseeable events that could occur over a span of 50 to 100 years or more, and which might induce the company to discontinue this operation in spite of guaranties given or promises made. If this were to happen, the Federal Government could not stand idly by and permit the public to remain unprotected. Instead, I would expect the AEC to step in and assume responsibility for the operation of this facility.

Such a step would make explicit something which has been an underlying assumption throughout, namely, that the Federal Government has the long term responsibility for the custody of radioactive waste material, even if the

waste involved was part of a privately owned and operated facility. This long term responsibility is natural in view of the fact that the activity which generated the waste material was licensed and even encouraged by AEC, and therefore this responsibility can be considered as inherent in any waste disposal operation which has a long term hazard associated with it. The question arises, in view of this long term responsibility on the part of the Federal Government, to what extent private industrial participation should be encouraged in this field. The AEC, in line with the language of the Atomic Energy Act, has consistently encouraged private industry to enter all phases of the nuclear industry; in fact, the AEC has committed itself to discontinue any service to private industry, if such service can be performed adequately by private industry. But encouragement of private industry participation in waste disposal activities would not really relieve the Federal Government of any responsibility in this field; it may merely postpone the time when the responsibility must be assumed by AEC. The question of industrial participation in the field of waste disposal, when stated realistically, is not whether AEC should turn over this field to private industry; it is rather whether encouragement of private industry's activity is worth the effort if the result is merely to secure private industry operation for some limited period. On that basis, it has been suggested that a private company might operate a waste disposal burial ground for 10 to 20 years and then turn it over to the Federal Government for future operation and custody.

While such an arrangement would meet some of the requirements of industry participation, it would accomplish this not without serious disadvantages. The first and most serious of these is one already mentioned,

namely, that private industry cannot permanently relieve the Federal Government of responsibility in this field and that, therefore, our efforts are merely for the sake of postponing the date when AEC would have to take over such facilities. Second, from the point of cost, it is likely that temporary private ownership and control may eventually result in higher Government costs since private ownership will probably continue as long as the facility produces significant revenues. The cost which the Government would later incur in operating and caring for abandoned facilities, might be relatively higher, since there would be little chance of contributions from users. Third, the Federal Government, in meeting its obligation of long term responsibility in this field, may at times be faced with operational problems of some complexity since it would be forced to take over facilities in whatever physical state they might be. Finally, in order to prevent the proliferation of radioactive burial grounds, some steps would have to be taken to limit the number of such sites.

Given these conflicting considerations on the subject of industrial participation in waste disposal, it is difficult to generalize and say definitely that such private participation will or will not be encouraged by the AEC. The AEC, is fully aware of the many companies and individuals who have expressed an interest to enter this field on a commercial basis, and some of them are already engaged in some phases of waste disposal, like sea disposal. But frankly, at the present state of knowledge, it is our current view that private ownership of burial sites is not in the public interest. We are particularly conscious of the importance of a waste disposal service being available, on a completely safe basis and at a reasonable cost, to all members of the atomic energy industry. We also believe that the assured existence of such a service can do much to stimulate a private atomic energy industry.

Waste Disposal for the Dresden Nuclear Power Station

By

T.G. LeClair, Manager of Research and Development, Commonwealth Edison Company

The hearings by the Joint Committee on the subject of waste disposal are most timely. They should furnish a perspective on the nature of atomic wastes and how they should be handled to avoid becoming a problem.

I assume that my invitation to present a statement to this Committee was based on my connection with the Dresden Nuclear Power Project, the largest all-nuclear power plant now under construction in this country. Its wastes should be typical of the wastes from most water cooled heterogeneous reactors.

As this Committee has been informed in testimony by Willis Gale, our Chairman, the Dresden plant is being constructed under a $45,000,000 fixed price contract by General Electric Company. Therefore, the full responsibility for the design and for the provision of all necessary means to dispose of waste are those of General Electric. We believe that General Electric has provided this plant with every reasonable safety feature so that there will be no question of any radioactive waste endangering either the operating personnel or the public.

I propose to discuss what Commonwealth is providing in the way of precautions and trained operating personnel. I would like, however, to do this against a brief background on the broad waste disposal problem which is by no means a new one to the electric utility industry.

Waste from All Activities

Advancing civilization brings with it an ever increasing problem of waste disposal. All of us are contributors to a greater or

lesser degree, whether it be from our backyard trash burners, our home heating plants, our automobile, commercial wastes, or from industrial processes. Even wind generated pollutants are man-made also, if you recognize that the open fields stem largely from man clearing away forests and plowing up the prairies. Certainly the fallout from atomic bomb tests is a man-made by-product.

All of this is part of the price we pay for progress. These wastes we have created are the by-products of the activities that support our economy and increase our standard of living. The general problem then, as I see it, is to keep these wastes at levels we can live with. It is necessary to appraise and reappraise the situation from time to time, and provide control measures to make certain that these pollutants are maintained at a tolerable and safe level. As an example, certain urban areas, because of the heavy concentration of industry and local atmospheric conditions, have had to take intelligently planned and coordinated steps to control air pollution in recent years. Among those communities which have taken very aggressive steps in this direction are Los Angeles, Pittsburgh, and Chicago. Certainly it is appropriate at this time to appraise the pollution potential of nuclear power installations.

Waste from Fuel is Not New

To see the atomic waste problem in its proper perspective, I would like to draw a few comparisons between the wastes from fossil-fueled and nuclear power plants. The wastes from conventional plants are in the form either of solids, liquids, or gases. These same forms of waste appear in the operation of nuclear power plants but the magnitude and the importance of the problems in the two types of plants are far different.

Fossil vs Nuclear Wastes

Solids. In the year 1956 the electric power industry consumed in fossil fuels (coal, oil and gas) the equivalent of about 45,000 train loads of coal - assuming one hundred 50-ton cars per train. The power produced was a little over 478 billion kilowatt-hours. About 8½% by weight of these fuels remained, after burning, as waste in the solid forms known as slag, fly ash, or cinders. This meant that the industry needed to dispose of 4,000 train loads of solid waste.

If the entire electrical output of this country had been obtained from heterogeneous reactor plants with a burnup of 10,000 megawatt days per ton of uranium used, the total amount of nuclear fuel throughput would have been less than one-half a train load. Practically all of this would remain contained in the spent fuel elements and as such would be considered solid waste material. Even so, the volume of waste from the nuclear fuel would have been less than one hundredth of one percent as much as from fossil fuels.

Let us take a closer look at the hazard features of waste. Wastes from fossil-fueled plants in the solid form are relatively inert and consequently the disposal problem is almost entirely one of bulk. The radioactive wastes, on the other hand, are extremely dangerous and it is important that every precaution be taken to insure that the public's safety be preserved. I believe that if we continually recognize this problem, we have nothing to fear from the solid waste disposal problem. The spent fuel will, in most cases, be chemically processed to separate plutonium and other by-products. Even if the by-products from the spent fuel are not all utilized for new processes, the volume is so small it should be perfectly

practicable to store the residue indefinitely in leak-proof vaults. Using Dresden as an example, a fraction of the site area would store the solid waste from 100 years of operation.

Liquids. Liquid wastes are not large in volume from either the fossil-fueled or nuclear-fueled power plants. The fossil-fueled plants produce only ordinary sewage and a relatively small amount of waste obtained from blowing down boilers, cleaning up equipment, and from the spent solutions used for regenerating demineralizer systems.

Liquid wastes from nuclear plants are also relatively small in volume but, because of the possibility that radioactive materials can be dangerous when dissolved, or suspended in water, it is essential that they be continuously monitored and either diluted to tolerable limits or concentrated so that they can be stored the same as the solid wastes.

Gases. Gaseous wastes present a very interesting contrast. Many people do not realize, because of the proper precautions having been used over the past half-centruy, that tremendous volumes of gaseous waste are produced from the burning of fossil fuels. One ton of coal will produce about 3 tons of carbon dioxide gas. The stacks of power plants in this country in the year 1956 released over 600 millions of tons of carbon dioxide into the atmosphere. Carbon dioxide is harmful to man if concentrated. On the other hand, when properly dispersed, it is the life-giving substance to the vegetable foods which we consume. Other gaseous wastes, such as water vapor, sulfur dioxide, and a variety of other chemicals again do not become a problem so long as they are properly diluted. The high stacks on modern power plants have proved to be the most successful means of dispersing gases.

The nuclear power plant, by contrast, produces an extremely small amount of gaseous waste, most of which is confined within the cladding of the fuel elements. The balance, even though potentially dangerous, is in such minute amounts that with proper monitoring and hold-up it can be dispersed into the atmosphere with perfect safety.

I draw these comparisons, or contrasts merely to give the Committee the full understanding that waste is not a new problem and that the volumes of waste from nuclear plants are infinitesimal compared with those from other industries. By recognizing that radioactive wastes are extremely hazardous and by exercising proper care in handling them, it is my feeling industry can multiply its needs for power many times without encountering an inherently serious disposal problem.

Handling Wastes at Dresden

Let us come directly to the Dresden project. While this 180,000 kilowatt project seems large today, I expect it to be a small beginning to a fast growing nuclear power industry.

At the present time the handling and disposition of radioactive wastes does not appear to present any insurmountable difficulties. The Dresden Station waste handling facilities' design features and operational procedures are given in the attached Appendix. Proper operation of these facilities will result in the release of radioactivity to the environs in amounts that will normally not exceed 10% of the AEC maximum permissible limits.

What about the responsibilities of Commonwealth Edison as the operator of the plant? First of all, there is an elaborate operator training program and licensing procedure. All those persons responsible for the operation of the plant will be trained to have an understanding of all materials in the plant and how to handle them.

In addition, the organization setup provides that one supervisor shall be responsible for the disposal of wastes and the checking of radioactivity conditions about the plant. Also, there will be at least one man on duty on every shift regularly checking radiation levels in areas where radioactive wastes could appear.

Surveys have already begun on the surrounding off-site area to determine the background levels of radiation and will cover at least a year's time before the plant goes in operation. Very thorough surveys will be made of the air, well and river waters, vegetation, soil, milk, and even of fish life to determine background levels. After the plant goes into operation, these surveys will be continued in order to have a double check on the monitoring equipment and its operation within the plant. In short, Commonwealth Edison is preparing fully to discharge its obligations and responsibilities on the treatment of the waste problem.

Dresden Station has been designed on the assumption that spent fuel will be sent to fuel processing plants. Handling at that point will be discussed by other authors. All other solid and concentrated liquid waste will be stored on the plant property indefinitely. The volume is estimated to be so small and the safety and security precautions so good, there should be no reason to fear any possible contamination to the plant personnel or the public. It is also believed that the hazards, if any, from storing on the site should be substantially less than the possible resulting hazards from shipping these wastes to some remote storage point elsewhere in the country.

Governmental Control

The Sanitary Water Board of the State of Illinois has required that we obtain a special permit to discharge any liquid

waste into the Illinois River. This permit was issued March 17, 1958 after that agency had examined in detail all the plans for disposal of any possibly radioactive wastes. The permit provides for definite requirements on radiation levels and dilution. Copies of the permit have been supplied to the Committee. The AEC retains detailed control over waste disposal practices as a part of its procedure under our application for a facility operating license. Our Hazards Summary Report outlines in detail in Section "G" the design of the radioactive waste disposal system. This report was submitted to the AEC on September 3, 1957, and is now on file in the Public Document Room.

The last section of the report which has been filed with AEC as a part of our application for a facility operating license is the Operating Procedures and Emergency Plan. Section "L" of this report is titled "Waste System Operation" and outlines the procedures which the Company will follow to insure that the release of the radioactive waste is always below the limit set by the AEC.

A repetition of a description of the design features is unnecessary here, but my overall opinion is that the long term waste disposal from Dresden will present fewer problems than waste disposal from a coal-fired plant of the same size.

Economic Comparison

A preliminary cost comparison between the waste handling facilities at Dresden and that at a conventional coal-fired generating station was made. The capital investment cost comparison did not present much of a problem but due to the many possible variables in the operation and maintenance of the Dresden waste handling system, the operation cost comparison did.

The investment cost of the fly ash and slag handling equipment of a conventional coal fuel plant of about the size of Dresden is approximately $5.50 per KW of capability. The following equipment was included in this cost figure: precipitators, fly ash removal and temporary storage facilities, and the slag and pyrite handling system. The best figure of the investment cost of the waste handling facilities being installed at Dresden is $2,400,000 or $13.35 per KW of capability. The above cost figure included the equipment listed below.

1. Waste collection system.
   Tanks, piping, pumps, instrumentation, concrete vaults and shielded piping tunnel.
2. Waste treatment and storage facilities, radwaste building and all equipment, waste storage vaults, and liquid waste hold-up tanks.
3. Ventilating and process gas systems stack, temporary gas storage lines, supply and exhaust fans, ductwork, and controls.
4. Monitoring equipment.
   Process radiation monitors, portable monitoring instruments, and environs monitoring instruments and facilities.

The above investment cost figures show that Dresden waste handling facilities cost 2.4 times that of a conventional coal-fired plant.

The operating cost of fly ash and slag handling at a conventional plant averages $0.16 per KW capability per year. This figure includes the cost of moving the products from their respective hoppers at the boiler to permanent yard storage or loading on cars

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for removal from plant. The best estimated operating cost figure for the Dresden waste handling points to $0.25 per KW of capability per year. This figure includes sampling and analysis, transfer of wastes from point of collection or source to radwaste building or burial vaults and final disposition. A pessimistic estimate of one maintenance man full time on day shift is also included.

If the environs monitoring program is considered part of the waste handling operating cost at Dresden the figure would be increased from $0.25 to $0.38 per KW of capability per year.

This comparison indicates that Dresden operating cost for waste handling will be about 2.4 times that of a conventional plant.

National Record

There is evidence that proper safety precautions have paid off during the 13 or more years of operation of the present atomic energy industry. As you know, these installations have been operated primarily by universities and industrial companies for AEC. The methods developed for the safe handling of radioactivity at laboratories, on research and experimental reactors, and in the operation of the plutonium production plants have resulted in an enviable safety record. Workers in these installations have averaged an exposure of less than 1/3 of the radiation allowed by strict safety standards. Also, in all these years of operation no member of the public is known to have suffered an overexposure to radiation as a result of living near atomic energy production or laboratory centers.

Management of future projects should be in the hands of responsible organizations who will be able to continue this safety record.

Conclusion

May I suggest the following conclusions from the material here presented:

1. Most wastes from heterogeneous nuclear power plants are contained in spent fuel to be processed in plants similar to those now successfully operating.
2. The design and operating precautions taken in the earliest nuclear power plants have resulted in adequate safety at comparatively high cost. It is hoped that operating experience will reduce these costs.
3. Wastes which cannot be safely dispersed should be stored at the site instead of attempting to ship them to a remote point, at least until experience indicates otherwise.
4. Nuclear wastes present no immediate cause for concern. The industry has an enviable safety record and is conscious of the need to continue it.

With normal development of nuclear power, waste disposal should be no more, and perhaps less, of a problem than wastes from fossil fuels.

APPENDIX

DRESDEN STATION

Design and Operation of Waste Systems

The basic objective of the design of and operating procedures set up for the Dresden waste handling system is that routine disposal of radioactive wastes must not result in exposure of any person on or off the plant premises to radiation in excess of the maximum permissible limits. Actually, sufficient conservatism has been included to assure that the average off-site exposure will be considerably below the permissible limits (about 10% of the maximum permissible limits).

In addition, the system must comply with AEC licensing regulations and the State of Illinois Sanitary Water Board regulations.

The radioactive waste to be handled at Dresden Station will be present in three forms: (1) solids, (2) liquids, and (3) gases.

Dresden Solid Wastes

The solids can be divided into two catagories - (1) those shipped from the site and (2) those retained on the property.

1. The spent fuel would be considered as solid waste. At the present time, it is estimated that on the average 129 fuel assemblies (about one-fourth the core loading) will be replaced every year. The 129 fuel assemblies weigh approximately 24 tons and will be shipped to the AEC Hanford site for reprocessing. The ultimate handling of the radioactive fission products is given in the Hanford presentation by Mr. H. Parker.

2. Facilities are provided on the site for burial of all other solid wastes in watertight concrete vaults. Eight hundred to

thirteen hundred cubic feet of solid wastes are expected each year. Six to nine hundred cubic feet of this amount will be contaminated ion-exchange resins from the condensate clean-up, reactor clean-up, and waste treating system demineralizers. The resins will be discharged from the demineralizer tanks remotely by sluicing to a stainless steel tank contained in an underground concrete vault. Leakage from the resin storage tank will be noted by a leak detector in the concrete vault sump.

The other two to four hundred cubic feet of solid waste will include rags, paper, water and gas filters, and unsalvagable equipment. This solid waste will be compressed by baling where possible and buried in a separate 1200 cubic foot underground concrete vault.

The estimated radioactivity of these solid wastes is from four to eight thousand curies per year. Practically all of this radioactivity is expected to be contained in the ion-exchange resins.

Liquid Wastes

The radioactive liquid wastes will be handled by one or, if needed, two consecutive procedures out of the four possible methods being designed and built into the waste disposal facilities. The four systems are:

1. Controlled discharge to the Illinois River via the cooling water discharge flume.
2. Natural radioactive decay and subsequent discharge to the Illinois River.
3. Filtration and subsequent demineralization.
4. Concentration and storage on the plant property.

The liquid wastes will be handled batchwise and the particular method used for each batch of waste will depend on the water quality and

radioactivity level of the waste as determined by analysis of sample.

The liquid wastes expected at the Dresden plant can be divided into two types: (1) that which can be safely discharged to the Illinois River and (2) that which will be retained on the Dresden site.

1. Approximately 1.1 million gallons of radioactive liquid waste will be discharged safely to the river each year. One million gallons of this amount will come from the secondary side of the secondary steam generators. The balance will be made up of laboratory and laundry waste solutions. The activity level of these liquid wastes will be low. These wastes will be mixed at a controlled rate with the condenser cooling water leaving the site so that the activity level will normally not exceed 10 per cent of the maximum permissible concentration (MPC) for the individual radioisotopes present.

The radioactivity concentration of these liquid wastes is expected to be very low and only about 1.6 curies of activity is expected to be discharged to the river per year.

2. All other radioactive liquids will be retained on the site. About 1.5 to 2 million gallons of low total solids and low radioactivity level water will be treated by filtration and demineralization and stored in a 200,000 gallon aluminum storage tank. This high quality water will be recycled in the reactor system as make-up water.

Other liquid wastes with high total solids and either low or high radioactivity levels will be neutralized if necessary and concentrated by evaporation. This type of liquid waste will include the regenerant solutions from the condensate demineralizer system and decontamination and laboratory waste solutions and is expected to amount

to 200,000 gallons per year. The concentrated solution will be stored in a steel tank in an underground concrete vault.

Approximately 1200 curies of radioactivity may be contained in this concentrated solution.

3. Gases

The radioactive gaseous wastes from Dresden will be handled and controlled by three methods: (1) dispersion by discharging from the 300 foot stack, (2) temporary hold-up to allow natural decay time and subsequent filtration before being discharged from the stack and (3) filtration before release from local stacks. The radioactivity concentration of the gases being discharged from the stack and from the air ejectors will be continuously monitored in the hold-up piping and in the stack as a means of controlling emission of waste gases from the plant.

Approximately 60,000 cfm of ventilating air from the reactor enclosure and the turbine building will be discharged from the 300 foot stack. This air is normally safe to discharge to the environs because the only radioactivity will be due to particulate matter such as dust.

The process waste gases amount to about 400 cfm and the radioactivity is present as O^{19}, N^{16}, N^{17}, and H^{3}. About 300 cfm are discharged from the gland seal condenser into hold-up piping before being discharged up the stack. The other 100 cfm will come from the main condenser air ejectors. This gas flow will be continuously monitored when discharged into similar hold-up piping and filtered before being discharged to the 300 foot stack. In the stack both of these waste gases will be mixed with the ventilating airs from the reactor enclosure and turbine building.

Approximately ten curies of activity are expected to be discharged from the stack daily from the above sources of gaseous waste.

Any fission gas release from the system would be due to a fuel element cladding failure. These fission gases would normally be discharged in the condenser air ejector gas flow and be identified by the continuous activity monitor on the gas flow. When the activity level of this gas flow reached 8×10^5 micro curies per second a valve at the discharge of the hold-up piping will automatically close resulting in the shutdown of the reactor through the high condenser pressure safety channel.

Ventilation air from other buildings on the site is expected to be approximately 40,000 cfm and will be filtered and monitored ahead of its discharge through local stacks. The radioactivity of this air is expected to be present as particulate matter which will be filtered out and no activity is expected to be discharged to the environs.

Correct operation of the waste handling and disposal facilities at the Dresden plant will be checked by an on-site and off-site environs monitoring program. This program has been initiated and will have been in operation about one year before full power operation of the plant to obtain background data.

By continuing this environs monitoring program during operation a good check on the effective operation of the waste handling facilities will be available.

RADIOACTIVE WASTE DISPOSAL PRACTICES

FOR

THE ATOMIC POWER EQUIPMENT DEPARTMENT

GENERAL ELECTRIC COMPANY

SAN JOSE, CALIFORNIA

by

E. A. Sadowski
J. M. Smith
R. C. Thorburn

ABSTRACT

A brief description is given of the sources of radioactive waste and methods of handling wastes by the Atomic Power Equipment Department of the General Electric Company. No serious problems in waste disposal from fuel manufacture, boiling water reactor operation or from the laboratory components have developed. The estimated quantities and costs are outlined along with a brief discussion of possible cost reductions.

INDUSTRIAL RADIOACTIVE WASTE DISPOSAL

The Atomic Power Equipment Department of General Electric Company, representing the Company's efforts in the development of commercial nuclear power, stresses a high degree of control for wastes originating at its own facilities and in power plants being designed and built for others.

The Department recognizes that safe methods for disposal of radioactive waste are an important responsibility of industries using radioactive material. Waste disposal programs must insure that solids and effluent gaseous and liquid concentrations are within the permissible limits established by the best available information for the constituents of the waste.

WASTE DIPOSAL METHODS

The Department's facilities are located at two principal sites in the San Francisco Bay area. The engineering and manufacturing facilities are located in San Jose, several miles south of the southern extremity of the San Francisco Bay. The laboratory facilities are located at Vallecitos in the hills some twenty miles northeast of San Jose. These sites include a very large variety of potential waste disposal problems; however, through careful design and established operating procedures, quantities of radioactive materials released to the environs are extremely limited.

In San Jose, the radioactive waste disposal problem is associated with uranium fuel development and manufacture. All liquid waste originating in the fuel facility and several small laboratories is received in a hold-up tank. Trace quantities of uranium contained in these waste streams are essentially removed by flocculation with either calcium or iron hydroxides. After precipitation and removal of the solids, the waste batch is sampled and analyzed for uranium content. Release to the municipal sewage system is permitted only when the concentration found by analysis is less than 10% of the public drinking water limit for uranium established by the United States Atomic Energy Commission.

All effluent air from radioactive materials areas in the San Jose site is discharged through high efficiency multiple stage filters to reduce the uranium concentration in the air below detectable levels. Solid wastes, which include filters, contaminated equipment, etc. are removed by a waste disposal firm licensed by the United States Atomic Energy Commission.

Samples of air, vegetation, soil and water of the environs surrounding the General Electric plant and from the San Jose City Sewage Treatment Plant are taken routinely to assure that no uranium has been dispersed.

The principal nuclear facilities currently in operation at the Vallecitos Laboratory site are:

(1) The Vallecitos Boiling Water Reactor
(2) The Experimental Physics Laboratory
(3) The Radioactive Materials Laboratory
(4) The General Electric Test Reactor

All waste water released from the various laboratory facilities which is contaminated with radioactive materials or is potentially contaminated is either

1) removed from the laboratory site by a United States Atomic Energy Commission licensed waste disposal contractor or 2) released into one of two retention basins for analysis preparatory to discharge into Vallecitos creek. The retention basins are of reinforced concrete construction, each of 50,000 gallon capacity. When one basin is filled, it is isolated from the system and a sample is analyzed for radioactivity content. When the quality of the water is found to be acceptable for discharge, it is released into the creek. The Laboratory Staff works closely with the Bay Area Regional Water Pollution Control Board Staff in ascertaining suitability for discharge. To date, no radioactive contamination from the Vallecitos operation has been released via this outlet.

In the Vallecitos Boiling Water Reactor, experimental fuels are irradiated under conditions which would be encountered in normal power plant use. The Pacific Gas & Electric Company has constructed and operates an associated turbine-generator to generate commercial electric power during the operation of the reactor. Sources of liquid waste from this facility include any necessary drainage from the boiling water reactor and the steam system itself, periodic replacement of recirculated cooling tower water, and other miscellaneous building drainage.

Wastes from the reactor system itself are purified by use of mixed bed demineralizers. In some cases, the activity of waste liquids can be reduced to essentially zero by short term decay storage.

The waste water system consists of the reactor enclosure and turbine area sumps, a hold-up sump, and the waste water filter and demineralizer. Waste from the reactor and turbine sumps flows into the main hold-up sump. A sample is taken at this point and analyzed. If treatment is required, it is circulated through the waste demineralizer and filter until the radioactivity is removed. Water may then be discharged to the concrete retention basins with other laboratory wastes for analysis and discharge to Vallecitos creek. Methods are also available to remove wastes to a tank truck for off-site disposal, but this has not been necessary to date since primary means of controlling contamination have been successful.

The condenser cooling water circulated through a cooling tower presents an interesting sidelight to the radioactive liquid waste disposal problem. Water coming to the laboratory from the Hetch Hetchy Aqueduct system contains trace quantities of naturally occurring radioactive material and fallout from weapons testing. In the use of this water in the cooling tower, a high degree of concentration of the salts in the water takes place. Thus, naturally occurring radioactivity in water can lead to concentrations which exceed the normal disposal limit. This problem is immediately apparent at a plant where wastes are tested for radioactive materials, but this particular phenomenon occurs at any place a cooling tower is operated.

In the Physics laboratory, radioactive liquid waste problems are minimal. In critical assemblies used primarily for testing reactor fuels, water is present as a moderator. This water is replaced infrequently, but its usage is such that essentially no radioactivity is induced. This is confirmed by sampling and analysis prior to release to a dual retention basin system. If the water were contaminated it would be removed by tank truck.

A principal use of the radioactive materials laboratory is to perform tests on irradiated fuel elements. Small quantities of highly radioactive liquid wastes are generated in this facility. Where the volume is small and the radiation level high, the liquid waste is solidified and handled subsequently as an item for solid waste diposal. Large volumes of radioactive wastes are received in a series of catch tanks in the laboratory basement. Currently this waste is placed in a tank trailer for removal from the site by a licensed contractor. Other miscellaneous waste streams from this laboratory, which should contain no radioactive material, are discharged to the concrete retention basin system and tested to assure safety.

The General Electric Test Reactor just began operations. Potential waste problems are similar to that of the boiling water reactor in that reactor water and cooling water are the chief potential sources for waste volume. Similar handling and treatment methods will be used. Liquid wastes from the plant, other than the cooling tower wastes, are drained or pumped to one of three retention tanks. These tanks have a 25,000 gallon capacity to provide retention for the maximum expected volume of liquid wastes. The storage or retention tanks are divided into a "non-usable" and a "re-usable" system. Radioactive and other possibly suspect drains discharge to the non-usable retention tank. The usable retention tanks take low-level radioactive overflow or scheduled drainage from the reactor and pool cooling systems when it has been proven that these liquids are not radioactive. Water from the usable tanks is returned to the demineralized water storage tank via the pool cleanup demineralizer.

General Electric has also supplied a number of open pool test reactors for various foreign governments and domestic universities. While we do not have such a facility at the Vallecitos Laboratory, we have established temporary provisions for testing cores of these reactors prior to shipment. Again, liquid waste problems are limited primarily to low level activity in reactor cooling water. Particularly in the case of the pool reactor, most of the significant activities have a relatively rapid radioactive decay.

All solid wastes generated by the Laboratory operations are carefully packaged. A waste disposal contractor, licensed by the United States Atomic Energy Commission, removes the wastes if the radiation levels are sufficiently low for safe transport over the highways.

Some solid wastes are too radioactive to remove from the site due to the unreasonably large amount of shielding which would be required during transportation. Therefore, a radioactive waste storage facility consisting of a horizontal tube arrangement shielded by concrete on the ends and earth fill on the top and sides was constructed. Waste is entered into the receiving end, and waste with reduced radioactivity, as a result of the natural decay process, is removed from the discharge end. Hold-up time for radioactive waste decay may require a few years. An associated storage area consisting of 2.5 feet diameter drainage pipes buried vertically in the soil is also available for storage of filled 55 gallon drums. After sufficient decay these materials are removed by the waste disposal contractor.

Air contaminants fall into two broad categories; particulate and gaseous. Where needed, decontamination of the air is effected through highly efficient multiple stage filtration which reduces the particulate contamination to low levels. Where necessary, radiogases which decay rapidly are retained in a tank for a short period of time until quantities of radioactivity are reduced to acceptable levels.

At all locations where air which may reasonably be expected to contain radioactive contamination is discharged from the laboratory buildings, the exhaust air is monitored continuously. If traces of contamination pass through the filters the ventilation system can be stopped until repairs are made. The monitors consist of two stages; one measures and records the accumulation of particulate and filterable radioactive material; the other measures and records concentrations of gaseous and non-filterable radioactive materials.

In addition, the Laboratory site is surrounded by four environmental monitoring stations. Each station continuously measures and records variance in the total radioactivity as detected by a Geiger counter, and also measures and records accumulation of radioactivity on a filter, through which atmospheric air is continuously drawn. These stations have not indicated radioactivity above normal background levels at the periphery of the Laboratory site as a result of Laboratory operations. This survey program is further supplemented by the radioanalysis of samples of soil, vegetation and water from the environs of the site.

The waste diposal systems outlined above provide the control which is necessary to meet health protection requirements. Continued maintenance of high health protection standards is expected in the future. No unusual or serious problems have been encountered and no new problems are anticipated that cannot be solved by usual engineering methods.

ECONOMICS OF WASTE PROGRAM

The San Jose facility, which manufactures reactor fuels, generates approximately 1500 gallons of potentially contaminated liquid wastes a week. In general, the contaminant consists of small suspended UO_2 particles from the laundry operations, floor mopping, etc. which are easily separated by the flocculating process. Chemicals for the operation cost less than $1/week and an additional two hours/week of laboratory technician's time is required for analyses. Excluding amortization of the floc tank and associated pumps and equipment, treatment costs about 5¢ a gallon including overhead costs. The flocculating equipment costs $7500 and with a six year amortization, the additional cost would be 1.6¢/gallon.

The solid waste volume at San Jose is approximately 800 cu ft/month and costs $6.10/cu ft. of volume for removal by the waste disposal contractor.

At Vallecitos, the radioactive materials cannot be economically separated from some of the liquid wastes, particularly those from the radioactive materials laboratory and associated chemical operations. These are the wastes that have been accumulated in tanks and are removed by the waste disposal contractor in truckload quantities. The average quantity of liquid waste removed during 1958 was 2300 gallons per month. The cost of liquid waste diposal ranges from 30¢ to 50¢ per gallon. The cost of storing the waste until removed is not included.

The average quantity of solid waste removed from Vallecitos during 1958 was 279 cubic feet per month and the cost of disposal varied from $3.25 to $6.10 per cubic foot. This cost does not include packaging and storage until disposal.

A total of 263 cubic feet of radioactive waste is currently stored at the Laboratory awaiting natural decay to lower levels to enable transportation without costly shielding. Decay to suitable levels for transport will vary from a few months to years.

Cost of shielded storage space for radioactive waste varies considerably with the radiation levels and nature of the waste. The minimum construction cost of shielded storage space at Vallecitos is $11 per cubic foot of usable space.

Future Estimates

The non-volatile characteristics of UO_2 dust at the San Jose facility offer attractive possibilities for incineration. Studies indicate that a suitable incinerator can be built for approximately $5000 to reduce the solid waste volume by an estimated 90%. Decontamination of the effluent gases will be effected by a cyclone separator backed up by a high efficiency, high temperature filter. Reclamation of U from the ashes may also prove feasible. A further reduction in the cost of liquid disposal from San Jose is anticipated through the installation of larger floc tanks to reduce the manhours required per gallon treated.

At Vallecitos waste volume reduction is complicated by the variety and the physical state of the radioisotopes being used. Studies of primary interest at the present time include the economics of mechanical waste compression, the feasibility of incineration of combustible solids and the possibility of solar evaporation of liquid wastes.

(At 1:10 p.m., Monday, February 2, 1959, a recess was taken until Tuesday, February 3, 1959, at 10 a.m.)

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